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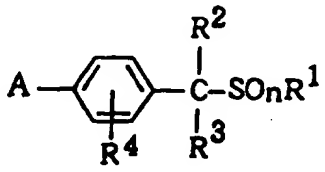
(54) Title: BENZYLSULFIDE DERIVATIVE, PROCESS FOR ITS PRODUCTION AND PESTICIDE

(57) Abstract

The present invention presents a benzylsulfide derivative of formula (I) or its salt: wherein R^1 is a C_{1-6} alkyl group, a C_{1-6} haloalkyl group, a C_{2-4} alkenyl group, a cyano group, etc., and each of R^2 and R^3 is a hydrogen atom, a halogen atom, a cyano group, a C_{1-4} alkyl group, a C_{1-3} haloalkyl group, etc., R^4 is a hydrogen atom, a halogen atom, a C_{1-4} alkyl group, etc., A is a hydrazinoaralkyl group or hydrazonoaralkyl group, and n is 0, 1 or 2; a process for its production; and a pesticide containing such a benzylsulfide derivative as an active ingredient. The benzylsufide derivative of the present invention is capable of controlling various pests without adversely affecting crop plants.



(I)

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DESCRIPTION

TITLE OF THE INVENTION

BENZYLSULFIDE DERIVATIVE, PROCESS FOR ITS PRODUCTION AND PESTICIDE

TECHNICAL FIELD

The present invention relates to a novel benzylsulfide derivative, a process for its production and a pesticide containing it as an active ingredient.

BACKGROUND ART

Heretofore, it has been reported, for example, in U.S. Patent 3,732,307 and Japanese Unexamined Patent Publications No. 122261/1979 and No. 45452/1981 that benzohydrazonophenylsulfide derivatives are useful as insecticides. However, the benzylsulfide derivative of the present invention has not been known.

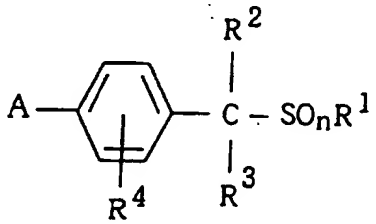
In recent years, some of conventional commercial insecticides have been restricted in their use in view of problems such as the residual effects, accumulation or environmental pollution, and some have become not so effective as the pests have acquired resistance during their use for a long period of time. Therefore, it has been desired to develop a new insecticide which is highly effective at a low dose and which is excellent in safety.

The present inventors have synthesized various benzylsulfide derivatives and have studied their physiological activities. As a result, it has been found

that the compound of the present invention exhibits outstanding pesticidal activities against various pests, particularly against agricultural and horticultural pests including lepidopteran injurious insects represented by diamond back (Plutella xylostella), Asiatic rice borer (Chio suppressalis) and beat armyworm (Spodoptera exigua), hemipteran injurious insects represented by brown planthopper (Nilaparvata lugens), green rice leafhopper (Nephotetlix cincticeps) and cotton aphid (Aphis gossypii) and elytron injurious insects represented by adzuki bean weevil (Callosobrunchus chinensis). The present invention has been accomplished on the basis of this discovery.

DISCLOSURE OF INVENTION

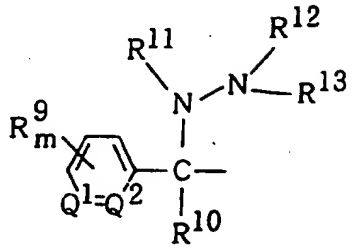
That is, the present invention provides (1) a benzylsulfide derivative of the formula (I) or its salt:



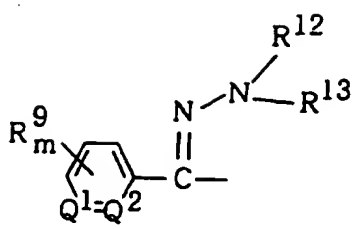
(I)

wherein R^1 is a C_{1-6} alkyl group, a C_{1-4} cyanoalkyl group, a C_{1-4} hydroxyalkyl group, a C_{3-6} cycloalkyl group, a C_{1-6} haloalkyl group, a C_{2-4} alkenyl group, a C_{2-4} alkynyl group, a phenyl group (which may be substituted by a halogen atom or a C_{1-4} alkyl group), a cyano group, a benzyl group (which may be substituted by a halogen atom), a thiazolyl group, a C_{1-4} alkylcarbamoyl group or

a group of the formula $-N(R^5)R^6$; each of R^2 and R^3 which are independent of each other, is a hydrogen atom, a halogen atom, a cyano group, a C_{1-4} alkyl group, a C_{1-3} haloalkyl group, a C_{1-4} alkylthio group, a C_{1-4} alkylcarbonyl group, a carboxyl group, or a C_{1-4} alkoxycarbonyl group; or R^2 and R^3 may form a 3- to 6-membered ring together with the carbon atom to which they are bonded; or R^1 and R^2 may form a 3- to 8-membered ring having one or more hetero atoms, together with the sulfur and carbon atoms to which they are respectively bonded; R^4 is a hydrogen atom, a halogen atom, a C_{1-4} alkyl group, a C_{1-4} haloalkyl group, a C_{1-4} alkoxy group or a C_{1-4} haloalkoxy group; each of R^5 and R^6 which are independent of each other, is a hydrogen atom, a C_{1-6} alkyl group or a C_{1-4} haloalkyl group; or R^5 and R^6 may together form a group of the formula $=CR^7R^8$; or R^5 and R^6 may form a 4- to 8-membered ring having one or more hetero atoms, together with the nitrogen atom to which they are bonded; R^7 is a hydrogen atom, a C_{1-3} alkyl group or a C_{1-3} alkylthio group; R^8 is a C_{1-3} alkylthio group or a C_{1-3} alkylamino group; or R^7 and R^8 may form a saturated or unsaturated 4- to 8-membered ring together with the carbon atom to which they are bonded; A is a hydrazinoaralkyl or hydrazonoaralkyl group of the formula (A1) or (A2):



[A1]



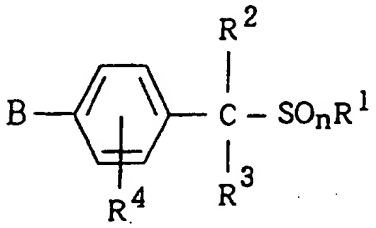
[A2]

R^9 is a hydrogen atom, a halogen atom, a nitro group, a cyano group, a C_{1-4} alkyl group, a C_{1-4} haloalkyl group, a C_{1-4} alkoxy group, a C_{1-4} haloalkoxy group, a C_{1-4} alkylthio group, a C_{1-4} haloalkylthio group, a C_{1-4} alkylsulfonyl group, a C_{2-4} alkylsulfonylmethyl group, a C_{1-4} haloalkylsulfonyloxy group, a phenyl group (which may be substituted by a halogen atom) or a phenoxy group (which may be substituted by a halogen atom); or two R^9 may together form a 5- or 6-membered ring; R^{10} is a hydrogen atom or a C_{1-4} alkyl group; each of R^{11}, R^{12} and R^{13} which are independent of one another, is a hydrogen atom, a cyano group, a C_{1-6} alkyl group, a C_{1-4} haloalkyl group, a C_{2-10} alkoxyalkyl group, a C_{3-8} alkoxyalkoxyalkyl group, a C_{2-6} alkylthioalkyl group, a C_{2-6} alkenyl group, a C_{2-4} alkynyl group, a C_{1-4} cyanoalkyl group, a benzyl group (which may be substituted by a halogen atom, a C_{1-4} haloalkyl group or a C_{1-4} alkyl group), a group of the formula $-COR^{14}$, a group of the formula $-CSR^{14}$, a group of the formula $-COOR^{15}$, a group of the formula $-COSR^{15}$, a group of the formula $-CON(R^{16})R^{17}$, a group of the formula $-CSN(R^{16})R^{17}$, a group of the formula $-SN(R^{18})R^{19}$, a group

of the formula $-SO_2R^{20}$ or a group of the formula $-C(R^{21})=CHR^{22}$; or R^{12} and R^{13} may together form a group of the formula $=CR^{23}R^{24}$; or R^{12} and R^{13} may form a 4- to 8-membered ring having one or more hetero atoms, together with the nitrogen atom to which they are bonded; R^{14} is a hydrogen atom, a C_{1-20} alkyl group, a C_{1-8} haloalkyl group, a C_{2-12} alkoxyalkyl group, a C_{2-10} haloalkoxyalkyl group, a C_{3-16} alkoxyalkoxyalkyl group, a C_{4-22} alkoxyalkoxyalkoxyalkyl group, a C_{2-6} alkylthioalkyl group, a C_{3-6} cycloalkyl group, a C_{1-5} hydroxyalkyl group, a C_{1-6} aminoalkyl group, a C_{1-6} amidoalkyl group, a C_{1-8} cyanoalkyl group, a C_{3-12} alkoxycarbonylalkyl group, a C_{2-6} alkenyl group, a C_{2-4} alkynyl group, a phenyl group (which may be substituted by a halogen atom, a nitro group, a C_{1-4} alkyl group, a C_{1-4} haloalkyl group, a phenoxy group or a C_{1-4} alkoxy group), a naphthyl group (which may be substituted by a halogen atom or a C_{1-4} alkyl group) or a hetero aromatic ring group (which may be substituted by a halogen atom or a C_{1-4} alkyl group); R^{15} is a C_{1-20} alkyl group, a C_{2-8} haloalkyl group, a C_{2-12} alkoxyalkyl group, a C_{2-6} alkenyl group, a C_{2-4} alkynyl group, a benzyl group (which may be substituted by a halogen atom, a C_{1-4} alkoxy group or a C_{1-4} alkyl group) or a phenyl group (which may be substituted by a halogen atom); R^{16} is a hydrogen atom or a C_{1-4} alkyl group; R^{17} is a hydrogen atom, a C_{1-6} alkyl group or a phenyl group (which may be substituted by a halogen atom, a C_{1-4}

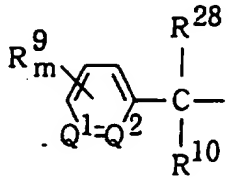
haloalkoxy group or a C_{1-4} alkyl group); each of R^{18} and R^{19} which are independent of each other, is a C_{1-4} alkyl group (which may be substituted by a C_{1-4} alkoxycarbonyl group), or a C_{2-5} alkoxyalkyl group; or R^{18} and R^{19} may form a 5- or 6-membered ring together with the nitrogen atom to which they are bonded; R^{20} is a C_{1-4} alkyl group, a C_{1-4} haloalkyl group or a C_{2-4} dialkylamino group; R^{21} is a hydrogen atom or a C_{1-6} alkyl group; R^{22} is a C_{2-4} acyl group or a C_{2-6} alkoxycarbonyl group; each of R^{23} and R^{24} which are independent of each other, is a hydrogen atom, a halogen atom, a C_{1-6} alkyl group or a group of the formula $-N(R^{25})R^{26}$; each of R^{25} and R^{26} which are independent of each other, is a hydrogen atom, a C_{1-4} alkyl group, a C_{1-4} alkoxy group, a C_{2-12} alkoxyalkyl group or a group of the formula $-SO_2R^{27}$; or R^{25} and R^{26} may form a 5- or 6-membered ring together with the nitrogen atom to which they are bonded; R^{27} is a C_{1-8} alkyl group or a phenyl group (which may be substituted by a halogen atom or a C_{1-4} alkyl group); each of Q^1 and Q^2 is a nitrogen atom or a group of the formula $-CR^9$; m is an integer of from 1 to 3; and n is 0, 1 or 2;

(2) a benzylsulfide derivative of the formula (II):

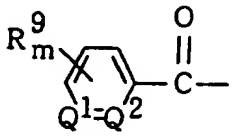


(II)

wherein R^1, R^2, R^3, R^4 and n are as defined in Claim 1; and B is an aralkyl or arylcarbonyl group of the formula (B1) or (B2):



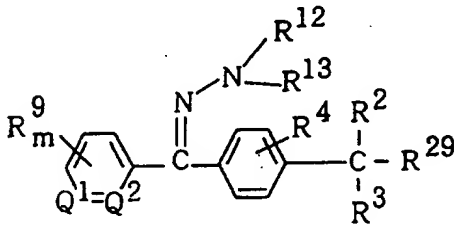
[B1]



[B2]

wherein R^9, R^{10}, m, Q^1 and Q^2 are as defined in Claim 1, and R^{28} is a halogen atom or a hydroxyl group;

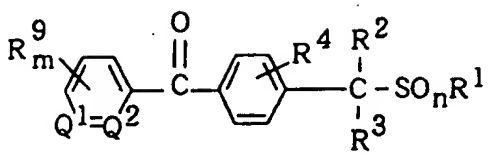
(3) a benzophenonehydrazone derivative of the formula (III):



(III)

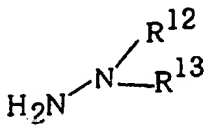
wherein R^4, R^9, R^{12}, R^{13}, m, Q^1 and Q^2 are as defined in Claim 1; each of R^2 and R^3 which are independent of each other, is a hydrogen atom or a C_{1-4} alkyl group, and R^{29} is a halogen atom, a mercapto group or a hydroxyl group;

(4) a process for producing a benzylsulfide derivative wherein A is a group of the formula (A2) as defined in Claim 1, which comprises reacting a compound of the formula (IV):



(IV)

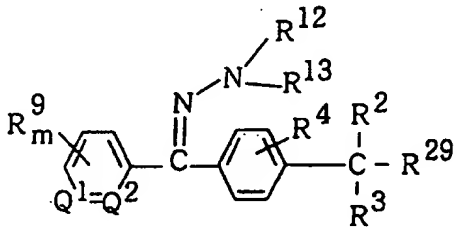
wherein R^1, R^2, R^3, R^4, R^9, m, n, Q^1 and Q^2 are as defined in Claim 1, with a compound of the formula (V1):



(V1)

wherein R^{12} and R^{13} are as defined in Claim 1;

(5) a process for producing a benzylsulfide derivative wherein A is a group of the formula (A2) as defined in Claim 1, which comprises reacting a compound of the formula (III);



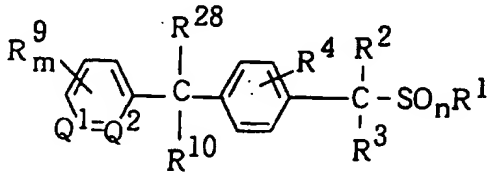
(III)

wherein R^2, R^3, R^4, R^9, R^{12}, R^{13}, R^{29}, m, Q^1 and Q^2 are as defined in Claim 3, with a compound of the formula (V2):

$Z-R^1$ (V2)

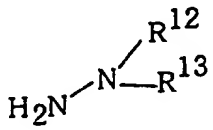
wherein Z is a halogen atom, a C_{1-4} alkylsulfonyl group or a benzenesulfonyloxy group (which may be substituted by a methyl group) when R^{29} is a mercapto group, or a group of the formula a $-S(O)_nM$ when R^{29} is a halogen atom, or a group of the formula $-SSR^1$ when R^{29} is a hydroxyl group; R^1 is a C_{1-6} alkyl group, a C_{1-4} cyanoalkyl group, a C_{3-6} cycloalkyl group, a C_{1-6} haloalkyl group, a C_{2-4} alkenyl group or a benzyl group (which may be substituted by a halogen atom); M is an alkali metal; and n is 0 or 2;

(6) a process for producing a benzylsulfide derivative wherein A is a group of the formula (A1) as defined in Claim 1, which comprises reacting a compound of the formula (VI):



(VI)

wherein R^1, R^2, R^3, R^4, R^9, R^{10}, m, n, Q^1 and Q^2 are as defined in Claim 1, and R^{28} is a halogen atom, with a compound of the formula (V1):



(V1)

wherein R^{12} and R^{13} are as defined in Claim 1; and

(7) a pesticide containing a benzylsulfide derivative

as defined in Claim 1, as an active ingredient.

In the present invention, the halogen atom represents a fluorine atom, a chlorine atom, a bromine atom or an iodine atom.

The alkyl group means a linear or branched C_{1-20} alkyl group such as a methyl group, an ethyl group, a n-propyl group, an isopropyl group, a n-butyl group, an isobutyl group, a sec-butyl group, a tert-butyl group, a n-pentyl group, an isoamyl group, a neopentyl group, a n-hexyl group, an isohexyl group, a 3,3-dimethylbutyl group, a n-heptyl group, a n-octyl group, a n-nonyl group or a n-decyl group.

The cycloalkyl group represents a C_{3-6} cycloalkyl group such as a cyclopropyl group, a cyclopentyl group or a cyclohexyl group.

The alkenyl group represents a linear or branched C_{1-6} alkenyl group such as an ethenyl group or a 2-propenyl group.

The haloalkyl group represents a linear or branched C_{1-8} alkyl group which is substituted from 1 to 10 halogen atoms which may be the same or different such as a chloromethyl group, a trifluoromethyl group or a tetrafluoroethyl group.

The cyanoalkyl group represents a linear or branched C_{1-8} alkyl group which is substituted by a cyano group.

The hydroxyalkyl group represents a linear or branched C_{1-8} alkyl group which is substituted by a

hydroxyl group.

The alkoxy group represents an alkyl-O- group wherein the alkyl moiety is as defined above, and it may, for example, be a methoxy group or an ethoxy group.

The haloalkoxy group represents a haloalkyl-O- group wherein the haloalkyl moiety is as defined above, and it may, for example, be a trifluoromethoxy group or a 2-chloroethoxy group.

The alkylthio group represents an alkyl-S- group wherein the alkyl moiety is as defined above, and it may, for example, be a methylthio group or an ethylthio group.

The haloalkylthio group represents a haloalkyl-S- group wherein the haloalkyl moiety is as defined above, and it may, for example, be a trifluoromethylthio group or a 2-chloroethylthio group.

The alkylsulfonyl group represents an alkyl-SO_2- group wherein the alkyl moiety is as defined above, and it may, for example, be a methylsulfonyl group or an ethylsulfonyl group.

The alkylsulfonylmethyl group represents an alkyl-SO_2CH_2- group wherein the alkyl moiety is as defined above, and it may, for example, be a methylsulfonylmethyl group or an ethylsulfonylmethyl group.

The alkylene group means a linear C_{1-8} alkylene group such as a methylene group, an ethylene group, a trimethylene group or a tetramethylene group.

The alkoxyalkyl group represents an alkyl-O-alkylene

group wherein the alkyl moiety and the alkylene moiety are as defined above, and it may, for example, be a methoxymethyl group or an ethoxymethyl group.

The alkylthioalkyl group represents an alkyl-S-alkylene group wherein the alkyl moiety and the alkylene moiety are as defined above, and it may, for example, be a methylthiomethyl group or an ethythiomethyl group.

The alkoxyalkoxyalkyl group represents an alkyl-O-alkylene-O-alkylene group wherein the alkyl moiety and each alkylene moiety are as defined above.

The alkoxyalkoxyalkoxyalkyl group represents an alkyl-O-alkylene-O-alkylene-O-alkylene group, wherein the alkyl and each alkylene group are as defined above.

The aminoalkyl group represents a linear or branched C_{1-8} alkyl group which is substituted by an amino group, a monoalkylamino group or a dialkylamino group.

The amidealkyl group represents a linear or branched C_{1-8} alkyl group which is substituted by an acylamino group or an N-alkyl-N-acylamino group.

The alkynyl group represents a linear C_{1-4} alkynyl group.

The hetero aromatic ring group represents a 5-membered aromatic ring group containing from 1 to 4 nitrogen, oxygen or sulfur atoms or a fused ring thereof with a benzene ring, or a 6-membered aromatic ring group containing from 1 to 3 nitrogen atoms or a fused ring thereof with a benzene ring, and it may, for example, be

a furyl group, a thienyl group, a pyrazolyl group, an imidazolyl group, a benzofuranyl group, a benzothiazolyl group, a pyridyl group, a pyrimidinyl group, a pyridazinyl group, a triazinyl group, a quinolyl group or a quinoxalinyl group.

In the compound of the present invention, the salt is a salt of the compound of the formula (I) with an acid, or a salt of the compound of the formula (I) wherein R^2 or R^3 is a carboxyl group, with a metal or an amine. The acid may, for example, be a hydrogen halide acid such as hydrochloric acid or hydrobromic acid, or a sulfonic acid such as methane sulfonic acid. The metal may, for example, be an alkali metal such as sodium or potassium, or an alkaline earth metal such as magnesium or calcium. The amine may, for example, be ammonia, isopropylamine or triethylamine.

A preferred group of compounds of the above formula (I) is a group of compounds wherein:

R^1 is a C_{1-4} alkyl group, a C_{1-2} cyanoalkyl group, a hydroxyethyl group, a cyclopentyl group, a C_{1-2} haloalkyl group, a phenyl group (which may be substituted by a halogen atom), a cyano group, a C_{1-4} alkylcarbamoyl group or a thiazolyl group;

each of R^2 and R^3 which are independent of each other, is a hydrogen atom, a methyl group or a C_{1-2} alkoxycarbonyl group; or R^1 and R^2 may form a 5-membered ring together with the sulfur and carbon atoms to which

they are respectively bonded;

R^4 is a hydrogen atom or a fluorine atom;

A is a hydrazinoaralkyl or hydrazonoaralkyl group of the formula (A1) or (A2);

R^9 is a hydrogen atom, a halogen atom, a nitro group, a cyano group, a methyl group, a trifluoromethyl group, a methoxy group, a C_{1-2} haloalkoxy group, a methylthio group, a difluoromethylthio group, a methylsulfonyl group, a methylsulfonylmethyl group, a trifluoromethylsulfonyloxy group, a phenyl group, a phenoxy group which may be substituted by a halogen atom, or a methylene dioxy group;

R^{10} is a hydrogen atom;

R^{11} is a hydrogen atom, a group of the formula $-COR^{14}$ or a group of the formula $-COOR^{15}$;

each of R^{12} and R^{13} which are independent of each other, is a hydrogen atom, a C_{1-4} alkyl group, a C_{1-4} haloalkyl group, a C_{2-10} alkoxyalkyl group, a C_{3-8} alkoxyalkoxyalkyl group, a C_{2-6} alkylthioalkyl group, a cyanomethyl group, a benzyl group (which may be substituted by a halogen atom or a trifluoromethyl group), a group of the formula $-COR^{14}$, a group of the formula $-COOR^{15}$, a group of the formula $-CONHR^{17}$, a group of the formula $-SO_2R^{20}$ or a group of the formula $-C(R^{21})=CHR^{22}$; or R^{12} and R^{13} may together form a group of the formula $=CR^{23}R^{24}$; or R^{12} and R^{13} may form a 5-membered ring together with the nitrogen atom to which they are

bonded;

R^{14} is a C_{1-10} alkyl group, a C_{1-4} haloalkyl group, a C_{2-6} alkoxyalkyl group, a C_{2-4} haloalkoxyalkyl group, a C_{3-10} alkoxyalkoxyalkyl group, a C_{4-12} alkoxyalkoxyalkoxyalkyl group, a cyclopropyl group, a C_{1-4} cyanoalkyl group, a C_{3-6} alkoxycarbonylalkyl group, a phenyl group (which may be substituted by a halogen atom, a nitro group, a C_{1-4} alkyl group, a trifluoromethyl group, a phenoxy group or a methoxy group), a naphthyl group, a pyridyl group, a thienyl group or a 2-furyl group;

R^{15} is a C_{1-10} alkyl group, a C_{2-6} haloalkyl group, a C_{2-6} alkoxyalkyl group or a phenyl group;

R^{16} is a hydrogen atom or a methyl group;

R^{17} is a hydrogen atom, a C_{1-6} alkyl group or a phenyl group (which may be substituted by a chlorine atom, a methyl group or a trifluoromethoxy group);

R^{20} is a methyl group or a trifluoromethyl group;

R^{21} is a hydrogen atom or a methyl group;

R^{22} is an acetyl group or a methoxycarbonyl group;

each of R^{23} and R^{24} which are independent of each other, is a hydrogen atom, a chlorine atom, a C_{1-4} alkyl group, a 1-triazolyl group or a group of the formula $-N(R^{25})R^{26}$;

each of R^{25} and R^{26} which are independent of each other, is a hydrogen atom, a C_{1-4} alkyl group, a methoxy group or a C_{2-4} alkoxyalkyl group;

R^{27} is a C_{1-4} alkyl group or a phenyl group (which may be substituted by a halogen atom or a methyl group);

each of Q^1 and Q^2 is a nitrogen atom or a group of the formula $-CR^9$;

m is an integer of 1 to 3; and

n is 0 when R^1 is a cyano group or a C_{1-4} alkylcarbamoyl group, or 0, 1 or 2 in other cases.

A preferred group of compounds of the above formula (II) may, for example, be a group of compounds wherein:

R^1 is a C_{1-4} alkyl group, a cyanomethyl group, a hydroxyethyl group, a cyclopentyl group, a C_{1-3} haloalkyl group, a phenyl group (which may be substituted by a halogen atom), a cyano group, a C_{1-4} alkylcarbamoly group or a thiazolyl group;

each of R^2 and R^3 which are independent of each other, is a hydrogen atom, a methyl group or a C_{1-2} alkoxycarbonyl group; or R^1 and R^2 may form a 5-membered ring together with the sulfur and carbon atoms to which they are respectively bonded;

R^4 is a hydrogen atom or a fluorine atom;

B is an aralkyl or arylcarbonyl group of the formula (B1) or (B2);

R^9 is a halogen atom, a trifluoromethyl group, a methoxy group, a C_{1-2} fluoroalkoxy group or a phenoxy group (which may be substituted by a halogen atom);

R^{10} is a hydrogen atom;

R^{28} is a chlorine atom or a hydroxyl group;

each of Q^1 and Q^2 is a nitrogen atom or a group of the formula $-CR^9$;

m is an integer of 1 or 2; and

n is 0 when R^1 is a cyano group or a C_{1-4} alkylcarbamoyl group, or 0, 1 or 2 in other cases.

A preferred group of compounds of the above formula (III) may, for example be a compound wherein:

each of R^2, R^3 and R^4 is a hydrogen atom;

R^9 is a chlorine atom substituted at the 4-position;

each of R^{12} and R^{13} is a hydrogen atom, a group of the formula $-COR^{14}$ or a group of the formula $-COOR^{15}$;

R^{14} is a C_{1-4} alkyl group;

R^{15} is a C_{1-4} alkyl group;

R^{29} is a chlorine atom, a mercapto group or a hydroxyl group;

each of Q^1 and Q^2 is a methine group; and

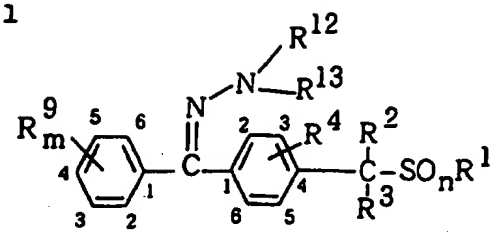
m is 1.

Now, typical specific examples of the compounds of the formulas (I), (II) and (III) of the present invention will be given in Tables 1 to 35. The compound numbers used in the tables will be referred to in the subsequent description.

The compound of the formula (I) has a C=N bond and accordingly has two geometrical isomers i.e. entgegen (E) isomer and zusammen (Z) isomer. As the compound of the present invention, the E isomer and the Z isomer may be used alone, or a mixture thereof may be employed.

Further, the compound of the formula (I) of the present invention may have tautonomers in some cases. For example, when the group of the formula $=CR^{23}R^{24}$ is represented by $=C(R^{23})-N(R^{25})R^{26}$, if R^{25} is a hydrogen atom, a compound having a partial structure of $-N=C(R^{23})NH-R^{26}$ will be present in an equilibrium state with a tautonomer having a partial structure of $-NH-C(R^{23})=N-R^{26}$. Accordingly, it should be understood that among the compounds of the present invention, those which are capable of having tautonomers, have such corresponding tautonomers even if such tautonomers are not specifically mentioned.

Table 1



Comp. No.	R^9_m	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})
I-1	4-Cl	CH_3	H	H	H	H	H	0	1.6503
I-2	4-Cl	CH_3	H	H	H	H	H	2	52- 54
I-3	4-Cl	CH_3	H	H	H	H	$C(CH_3)=NSO_2CH_3$	2	
I-4	4-Cl	CH_3	H	H	H	H	$CH=NSO_2CH_3$	2	
I-5	4-Cl	CH_3	H	H	H	H	$CH=NSO_2C_4H_9$	2	
I-6	4-Cl	CH_3	H	H	H	H	$C(C_2H_5)=NSO_2CH_3$	0	
I-7	4-Cl	CH_3	H	H	H	H	$C(C_2H_5)=NSO_2CH_3$	1	
I-8	4-Cl	CH_3	H	H	H	H	$C(C_2H_5)=NSO_2CH_3$	2	
I-9	4-Cl	CH_3	H	H	H	CH_3	$CH=NSO_2CH_3$	2	
I-10	4-Cl	CH_3	H	H	H	$COCH_3$	H	2	214-217
I-11	4-Cl	CH_3	H	H	H	COC_2H_5	H	0	91- 93
I-12	4-Cl	CH_3	H	H	H	COC_2H_5	CH_3	0	1.6319
I-13	4-Cl	CH_3	H	H	H	COC_2H_5	CH_3	1	141-143
I-14	4-Cl	CH_3	H	H	H	COC_2H_5	H	1	153-156
I-15	4-Cl	CH_3	H	H	H	COC_2H_5	H	2	159-160
I-16	4-Cl	CH_3	H	H	H	COC_3H_7	H	2	173-175
I-17	4-Cl	CH_3	H	H	H	COC_4H_9	H	2	147-149
I-18	4-Cl	CH_3	H	H	H	COC_5H_{11}	H	0	93- 96
I-19	4-Cl	CH_3	H	H	H	COC_5H_{11}	CH_3	0	1.6097
I-20	4-Cl	CH_3	H	H	H	COC_5H_{11}	CH_3	1	93-96
I-21	4-Cl	CH_3	H	H	H	COC_5H_{11}	H	1	1.6009
I-22	4-Cl	CH_3	H	H	H	COC_5H_{11}	H	2	110-113
I-23	4-Cl	CH_3	H	H	H	COC_5H_{11}	CH_3	2	115-117
I-24	4-Cl	CH_3	H	H	H	$CHOC_5H_{11}$	$CH_2OC_2H_5$	2	1.5722
I-25	4-Cl	CH_3	H	H	H	COC_6H_{13}	H	2	127-129
I-26	4-Cl	CH_3	H	H	H	COC_8H_{17}	H	2	116-118

Table 2

Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})
I-27	4-Cl	CH_3	H	H	H	$COCF_3$	H	2	
I-28	4-Cl	CH_3	H	H	H	CO-Ph	H	2	
I-29	4-Cl	CH_3	H	H	H	CO-(2-F,4-F)Ph	H	2	
I-30	4-Cl	CH_3	H	H	H	CO-(2-Cl,4-Cl,5-Cl)Ph	H	2	
I-31	4-Cl	CH_3	H	H	H	CO-(4-Cl)Ph	H	2	174-176
I-32	4-Cl	CH_3	H	H	H	CO-cyclopropyl	H	2	209-211
I-33	4-Cl	CH_3	H	H	H	CO-cyclopropyl	H	0	
I-34	4-Cl	CH_3	H	H	H	$COCH_2CH_2CH_2Cl$	H	2	180-182
I-35	4-Cl	CH_3	H	H	H	$COCH_2COOC_2H_5$	H	0	1.6149
I-36	4-Cl	CH_3	H	H	H	$COCH_2CH_2OH$	H	2	
I-37	4-Cl	CH_3	H	H	H	$COCH_2CN$	H	2	
I-38	4-Cl	CH_3	H	H	H	$COCH_2OCH_2CF_3$	H	2	
I-39	4-Cl	CH_3	H	H	H	$COCH_2OCH_3$	H	2	
I-40	4-Cl	CH_3	H	H	H	$CONH_2$	H	0	Unmeasurable
I-41	4-Cl	CH_3	H	H	H	$CONH_2$	H	1	195-197
I-42	4-Cl	CH_3	H	H	H	$CONH_2$	H	2	189-191
I-43	4-Cl	CH_3	H	H	H	COO-Ph	H	2	94- 96
I-44	4-Cl	CH_3	H	H	H	$COOC_2H_5$	H	2	164-166
I-45	4-Cl	CH_3	H	H	H	$COOC_2H_5$	H	0	1.6148
I-46	4-Cl	CH_3	H	H	H	$COOC_2H_5$	H	1	41- 43
I-47	4-Cl	CH_3	H	H	H	$COOC_2H_5$	CH_3	0	1.6042
I-48	4-Cl	CH_3	CH_3	H	H	$COOC_2H_5$	H	2	162-165
I-49	4-Cl	CH_3	CH_3	CH_3	H	$COOC_2H_5$	H	2	

Table 3

Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})
I-50	4-Cl	CH_3	H	H	H	$COOC_2H_5$	$CH_2OC_2H_5$	2	125-128
I-51	4-Cl	CH_3	H	H	H	$COOC_2H_5$	$CH_2OC_2H_5$	0	1.5853
I-52	4-Cl	CH_3	H	H	H	$COOC_3H_7$	H	0	1.6152
I-53	4-Cl	CH_3	H	H	H	$COOC_3H_7$	H	1	43- 45
I-54	4-Cl	CH_3	H	H	H	$COOC_3H_7$	H	2	167-169
I-55	4-Cl	CH_3	H	H	H	$COOC_3H_7$-i	H	2	159-160
I-56	4-Cl	CH_3	H	H	H	$COOC_4H_9$	H	2	68- 70
I-57	4-Cl	CH_3	H	H	H	$COOC_4H_9$-t	H	0	132-134
I-58	4-Cl	CH_3	H	H	H	$COOC_4H_9$-t	H	1	89- 93
I-59	4-Cl	CH_3	H	H	H	$COOC_4H_9$-t	H	2	193-195
I-60	4-Cl	CH_3	H	H	H	$COOCH_2CH_2Cl$	H	2	65- 67
I-61	4-Cl	CH_3	H	H	H	$COOCH_2CH_2OC_2H_5$	H	0	1.5822
I-62	4-Cl	CH_3	H	H	H	$COOCH_2CH_2OC_2H_5$	H	1	56- 58
I-63	4-Cl	CH_3	H	H	H	$COOCH_2CH_2OC_2H_5$	H	2	47- 49
I-64	4-Cl	CH_3	H	H	H	$COOCH_2CH_2OCH_3$	H	0	1.6179
I-65	4-Cl	CH_3	H	H	H	$COOCH_2CH_2OCH_3$	H	1	63- 65
I-66	4-Cl	CH_3	H	H	H	$COOCH_2CH_2OCH_3$	H	2	70- 72
I-67	4-Cl	CH_3	H	H	H	$COOCH_3$	H	0	40-4 2
I-68	4-Cl	CH_3	H	H	H	$COOCH_3$	H	1	176-177
I-69	4-Cl	CH_3	H	H	H	$COOCH_3$	H	2	197-199
I-70	4-Cl	CH_3	H	H	H	$COOCH_3$	CH_3	0	1.6238
I-71	4-Cl	CH_3	H	H	H	$COOCH_3$	CHF_2	0	1.5888
I-72	4-Cl	CH_3	H	H	H	$COOCH_3$	CH_3	1	1.6082
I-73	4-Cl	CH_3	H	H	H	$COOCH_3$	$CH_2OC_2H_5$	0	1.5911
I-74	4-Cl	CH_3	H	H	H	$COOCH_3$	CH_2SCH_3	0	1.6187
I-75	4-Cl	CH_3	H	H	H	$COOCH_3$	$CH_2OC_2H_5$	1	1.5949
I-76	4-Cl	CH_3	H	H	H	$COOCH_3$	$CH_2OC_2H_5$	2	61- 63
I-77	4-Cl	CH_3	H	H	H	$COOCH_3$	CH_3	2	64- 66
I-78	4-Cl	CH_3	H	H	H	$COOCH_3$	CH_2-  -Cl	0	1.6199
I-79	4-Cl	CH_3	H	H	H	$SO_2CH_2CF_3$	H	2	
I-80	4-Cl	CH_3	H	H	H	SO_2CH_2Cl	H	2	

Table 4

Comp. No.	R^9_m	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})
I-81	4-Cl	C_2H_5	H	H	H	H	H	2	45- 47
I-82	4-Cl	C_2H_5	H	H	H	$C(C_2H_5)=NSO_2CH_3$	H	0	1.6358
I-83	4-Cl	C_2H_5	H	H	H	$C(C_2H_5)=NSO_2CH_3$	H	1	45- 47
I-84	4-Cl	C_2H_5	H	H	H	$C(C_2H_5)=NSO_2CH_3$	H	2	180-181
I-85	4-Cl	C_2H_5	H	H	H	$C(C_2H_5)=NSO_2CH_2CH_3$	H	0	
I-86	4-Cl	C_2H_5	H	H	H	$C(C_2H_5)=NSO_2CH_2CH_3$	H	1	
I-87	4-Cl	C_2H_5	H	H	H	$C(C_2H_5)=NSO_2CH_2CH_3$	H	2	
I-88	4-Cl	C_2H_5	H	H	H	$COCH_3$	H	2	193-195
I-89	4-Cl	C_2H_5	H	H	H	COC_2H_5	H	0	1.6025
I-90	4-Cl	C_2H_5	H	H	H	COC_2H_5	H	1	49- 51
I-91	4-Cl	C_2H_5	H	H	H	COC_2H_5	H	2	122-125
I-92	4-Cl	C_2H_5	H	H	H	COC_5H_{11}	H	0	43- 45
I-93	4-Cl	C_2H_5	H	H	H	COC_5H_{11}	H	1	98-100
I-94	4-Cl	C_2H_5	H	H	H	COC_5H_{11}	H	2	105-107
I-95	4-Cl	C_2H_5	H	H	H	$COCH_2COOC_2H_5$	H	2	1.5988
I-96	4-Cl	C_2H_5	H	H	H	$COOCH_3$	H	0	1.6269
I-97	4-Cl	C_2H_5	H	H	H	$COOCH_3$	H	1	145-147
I-98	4-Cl	C_2H_5	H	H	H	$COOCH_3$	H	2	160-162
I-99	4-Cl	C_2H_5	H	H	H	$COOCH_3$	CH_3	0	1.6113
I-100	4-Cl	C_2H_5	H	H	H	$COOCH_3$	CH_3	1	1.6059
I-101	4-Cl	C_2H_5	H	H	H	$COOCH_3$	CH_3	2	1.5996
I-102	4-Cl	C_2H_5	H	H	H	$COOCH_3$	CHF_2	0	1.5838
I-103	4-Cl	C_2H_5	H	H	H	$COOCH_3$	CHF_2	1	
I-104	4-Cl	C_2H_5	H	H	H	$COOCH_3$	CHF_2	2	
I-105	4-Cl	C_2H_5	H	H	H	$COOCH_3$	$COOCH_3$	2	
I-106	4-Cl	C_2H_5	H	H	H	$COOCH_3$	$COOCH_3$	0	1.5988
I-107	4-Cl	C_2H_5	H	H	H	$COOCH_3$	$CONH_2$	2	
I-108	4-Cl	C_2H_5	H	H	H	$COOCH_3$	C_2H_5	2	
I-109	4-Cl	C_2H_5	H	H	H	$COOCH_3$	CH_2OCH_3	2	
I-110	4-Cl	C_2H_5	H	H	H	$COOCH_3$	COC_2H_5	2	
I-111	4-Cl	C_2H_5	H	H	H	$COOCH_3$	CH_2SCH_3	2	

Table 5

Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})
I-112	4-Cl	C_2H_5	H	H	H	$COOCH_3$	CH_2-⬡	2	
I-113	4-Cl	C_2H_5	H	H	H	$COOCH_3$	CHF_2	2	
I-114	4-Cl	C_2H_5	H	H	H	$COOC_2H_5$	H	0	1.6198
I-115	4-Cl	C_2H_5	H	H	H	$COOC_2H_5$	H	1	58- 61
I-116	4-Cl	C_2H_5	H	H	H	$COOC_2H_5$	H	2	68- 70
I-117	4-Cl	C_2H_5	H	H	H	$COOC_3H_7$	H	2	152-155
I-118	4-Cl	C_2H_5	H	H	H	$COOC_4H_9$	H	2	127-130
I-119	4-Cl	C_2H_5	H	H	H	$COOC_4H_9$-t	H	2	173-176
I-120	4-Cl	C_2H_5	H	H	H	$COOC_4H_9$-t	H	1	
I-121	4-Cl	C_2H_5	H	H	H	$COOCH_2CH_2OC_2H_5$	H	2	1.5748
I-122	4-Cl	C_2H_5	H	H	H	SO_2CH_3	H	2	86- 88
I-123	4-Cl	C_3H_7	H	H	H	$COCH_3$	H	2	
I-124	4-Cl	C_3H_7	H	H	H	COC_2H_5	H	2	120-122
I-125	4-Cl	C_3H_7	H	H	H	COC_5H_{11}	H	2	116-117
I-126	4-Cl	C_3H_7	H	H	H	$COOC_2H_5$	H	0	
I-127	4-Cl	C_3H_7	H	H	H	$COOC_2H_5$	H	1	
I-128	4-Cl	C_3H_7	H	H	H	$COOC_2H_5$	H	2	132-134
I-129	4-Cl	C_3H_7	H	H	H	H	H	2	88- 90
I-130	4-Cl	C_3H_7-i	H	H	H	COC_2H_5	H	2	130-132
I-131	4-Cl	C_3H_7-i	H	H	H	$COOC_2H_5$	H	2	
I-132	4-Cl	C_3H_7-i	H	H	H	$COOC_2H_5$	H	2	82- 84
I-133	4-Cl	C_3H_7-i	H	H	H	H	H	2	68- 70
I-134	4-Cl	C_4H_9	H	H	H	$COOC_2H_5$	H	2	114-116
I-135	4-Cl	CF_3	H	H	H	H	H	0	1.5969
I-136	4-Cl	CF_3	H	H	H	H	H	2	1.5871
I-137	4-Cl	CF_3	H	H	H	$CH=NSO_2CH_3$	H	2	63- 65
I-138	4-Cl	CF_3	H	H	H	$CH=NSO_2$-⬡	H	2	74- 78
I-139	4-Cl	CF_3	H	H	H	$CH=NSO_2CH_3$	H	0	

Table 6

Comp. No.	R^9_m	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})
I-140	4-Cl	CF_3	H	H	H	$C(CH_3)=CHCOCH_3$	H	2	121-123
I-141	4-Cl	CF_3	H	H	H	$C(CH_3)=CHCOOCH_3$	H	2	96- 98
I-142	4-Cl	CF_3	H	H	H	$CH=CHCOCH_3$	H	2	
I-143	4-Cl	CF_3	H	H	H	$CH=CHCOCH_3$	H	1	
I-144	4-Cl	CF_3	H	H	H	$C(C_2H_5)=NSO_2CH_3$	H	0	
I-145	4-Cl	CF_3	H	H	H	$C(C_2H_5)=NSO_2CH_3$	H	1	
I-146	4-Cl	CF_3	H	H	H	$C(C_2H_5)=NSO_2CH_3$	H	2	
I-147	4-Cl	CF_3	H	H	H	CH_2CF_3	H	0	
I-148	4-Cl	CF_3	H	H	H	CH_2CF_3	H	2	1.5539
I-149	4-Cl	CF_3	H	H	H	COC_2H_5	H	2	130-132
I-150	4-Cl	CF_3	H	H	H	COC_2H_5	H	0	121-123
I-151	4-Cl	CF_3	H	H	H	COC_2H_5	H	1	152-154
I-152	4-Cl	CF_3	H	H	H	COC_2H_5	H	2	
I-153	4-Cl	CF_3	H	H	H	COC_3H_7	H	2	125-126
I-154	4-Cl	CF_3	H	H	H	COC_4H_9	H	0	59- 61
I-155	4-Cl	CF_3	H	H	H	COC_4H_9	H	2	153-156
I-156	4-Cl	CF_3	H	H	H	COC_5H_{11}	H	2	122-124
I-157	4-Cl	CF_3	H	H	H	$COCF_3$	H	2	54- 56
I-158	4-Cl	CF_3	H	H	H	$COCF_3$	H	0	1.5659
I-159	4-Cl	CF_3	H	H	H	$COCH_2OC_2H_4OC_2H_4OC_2H_5$	H	2	78- 80
I-160	4-Cl	CF_3	H	H	H	$COCH_2OC_2H_4OC_2H_5$	H	2	101-103
I-161	4-Cl	CF_3	H	H	H	$COCH_2OC_2H_5$	H	2	126-128
I-162	4-Cl	CF_3	H	H	H	$COCH_2OCH_2CF_3$	H	2	120-122
I-163	4-Cl	CF_3	H	H	H	$COCH_3$	H	0	1.5972
I-164	4-Cl	CF_3	H	H	H	$COCH_3$	H	1	129-130
I-165	4-Cl	CF_3	H	H	H	$COCH_3$	H	2	112-114
I-166	4-Cl	CF_3	H	H	H	CONH-	H	2	81- 83
I-167	4-Cl	CF_3	H	H	H	CONH- 	H	2	

Table 7

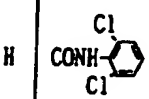
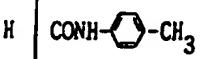
Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{25})
I-168	4-Cl	CF_3	H	H	H		H	2	132-134
I-169	4-Cl	CF_3	H	H	H		H	2	193-195
I-170	4-Cl	CF_3	H	H	H	$CONH_2$	H	2	
I-171	4-Cl	CF_3	H	H	H	$CONHC_4H_9$	H	2	179-181
I-172	4-Cl	CF_3	H	H	H	$CONHCH_3$	H	2	
I-173	4-Cl	CF_3	H	H	H	$COOC_2H_5$	H	0	75- 76
I-174	4-Cl	CF_3	H	H	H	$COOC_2H_5$	H	1	178-180
I-175	4-Cl	CF_3	H	H	H	$COOC_2H_5$	H	2	148-150
I-176	4-Cl	CF_3	H	H	H	$COOCH_3$	H	0	1.5921
I-177	4-Cl	CF_3	H	H	H	$COOCH_3$	H	1	181-183
I-178	4-Cl	CF_3	H	H	H	$COOCH_3$	H	2	151-153
I-179	4-Cl	CF_3	H	H	H	$COOCH_3$	CH_3	0	1.5802
I-180	4-Cl	CF_3	H	H	H	$COOCH_3$	CH_3	1	1.5820
I-181	4-Cl	CF_3	H	H	H	SO_2CF_3	H	2	32- 34
I-182	4-Cl	CF_3	H	H	H	SO_2CH_3	H	2	64- 65
I-183	4-Cl	CHF_2	H	H	H	COC_2H_5	H	0	1.6203
I-184	4-Cl	CHF_2	H	H	H	COC_2H_5	H	2	158-160
I-185	4-Cl	CHF_2	H	H	H	$COOC_2H_5$	H	2	197-199
I-186	4-Cl	CHF_2	H	H	H	$COOCH_3$	CH_3	0	1.5981
I-187	4-Cl	CHF_2	H	H	H	$COOCH_3$	H	0	1.6213
I-188	4-Cl	CHF_2	H	H	H	$COOCH_3$	H	1	71- 73
I-189	4-Cl	CHF_2	H	H	H	$COOCH_3$	H	2	171-173
I-190	4-Cl	CHF_2	H	H	H	H	H	0	1.6273
I-191	4-Cl	C_2F_5	H	H	H	$COOCH_3$	H	2	166-168
I-192	4-Cl	CF_2CHF_2	H	H	H	$COOCH_3$	H	0	1.5801
I-193	4-Cl	C_2F_5	H	H	H	$COOCH_3$	H	0	1.5649
I-194	4-Cl	C_2F_5	H	H	H	$COOC_2H_5$	H	0	1.5629

Table 8

Comp. No.	R^9_m	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})
I-195	4-Cl	C_2F_5	H	H	H	$COOC_2H_5$	H	2	94-97
I-196	4-Cl	CF_2CHF_2	H	H	H	$COOC_2H_5$	H	2	136-137
I-197	4-Cl	CF_2CHF_2	H	H	H	$COOCH_3$	H	2	145-148
I-198	4-Cl	C_3F_7	H	H	H	$COOC_2H_5$	H	2	
I-199	4-Cl	C_3F_7-i	H	H	H	$COOC_2H_5$	H	2	
I-200	4-Cl	CF_2CHFCF_3	H	H	H	$COOC_2H_5$	H	0	
I-201	4-Cl	CF_2CHFCF_3	H	H	H	$COOC_2H_5$	H	1	
I-202	4-Cl	CF_2CHFCF_3	H	H	H	$COOC_2H_5$	H	2	
I-203	4-Cl	CH_2- 	H	H	H	$COOC_2H_5$	H	0	
I-204	4-Cl	CH_2Br	H	H	H	COC_2H_5	H	2	
I-205	4-Cl	CH_2Br	H	H	H	$COOC_2H_5$	H	2	
I-206	4-Cl	CH_2CF_3	H	H	H	$COOC_2H_5$	H	2	Unmeasurable
I-207	4-Cl	CH_2CF_3	H	H	H	$COOCH_3$	H	2	154-156
I-208	4-Cl	$CHCl_2$	H	H	H	$COOC_2H_5$	H	2	
I-209	4-Cl	CH_2CH_2OH	H	H	H	$COOC_2H_5$	H	0	1.6141
I-210	4-Cl	CH_2CH_2OH	H	H	H	$COOC_2H_5$	H	2	75- 78
I-211	4-Cl	CH_2Cl	H	H	H	CO- 	H	2	176-178
I-212	4-Cl	CH_2Cl	H	H	H	COC_2H_5	H	2	63- 65
I-213	4-Cl	CH_2Cl	H	H	H	COC_3H_7	H	2	112-114
I-214	4-Cl	CH_2Cl	H	H	H	COC_5H_{11}	H	2	91- 93
I-215	4-Cl	CH_2Cl	H	H	H	$COCH_2Cl$	H	2	180-181
I-216	4-Cl	CH_2Cl	H	H	H	$COCH_2CN$	H	2	68- 70
I-217	4-Cl	CH_2Cl	H	H	H	$COCH_3$	H	2	185-187
I-218	4-Cl	CH_2Cl	H	H	H	$COOC_2H_5$	H	2	166-168
I-219	4-Cl	CH_2Cl	H	H	H	$COOC_3H_7$	H	2	
I-220	4-Cl	CH_2Cl	H	H	H	$COOCH_2CH_2OCH_3$	H	2	56- 58
I-221	4-Cl	CH_2Cl	H	H	H	$COOCH_2CH_2OC_2H_5$	H	2	60- 62
I-222	4-Cl	CH_2Cl	H	H	H	$COOC_3H_7$	H	2	105-107

Table 9

Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})
I-223	4-Cl	CH_2Cl	H	H	H	$COOCH_3$	H	2	160-162
I-224	4-Cl	CH_2Cl	H	H	H	H	H	2	1.6258
I-225	4-Cl	CH_2CN	H	H	H	$COOC_2H_5$	H	0	89- 91
I-226	4-Cl	CH_2CN	H	H	H	$COOC_2H_5$	H	2	81- 83
I-227	4-Cl	CH_2CH_2CN	H	H	H	$COOC_2H_5$	H	0	
I-228	4-Cl	CH_2CN	H	H	H	$COOC_2H_5$	H	1	
I-229	4-Cl	CH_2CH_2CN	H	H	H	$COOC_2H_5$	H	1	
I-230	4-Cl	CH_2CH_2CN	H	H	H	$COOC_2H_5$	H	2	
I-231	4-Cl	CH_2-  -Cl	H	H	H	$COOC_2H_5$	H	0	
I-232	4-Cl	CN	H	H	H	$COOC_2H_5$	H	0	1.6228
I-233	4-Cl	CN	H	H	H	$COOC_2H_5$	H	1	
I-234	4-Cl	-  Cl	H	H	H	$COOC_2H_5$	H	2	
I-235	4-Cl	-  -CH_3	H	H	H	$COOC_2H_5$	H	2	176-179
I-236	4-Cl	-  -CH_3	H	H	H	$COOC_2H_5$	H	1	
I-237	4-Cl		H	H	H	$COOC_2H_5$	H	2	
I-238	4-Cl		H	H	H	$COOC_2H_5$	H	0	1.6524
I-239	4-Cl		H	H	H	$COOC_2H_5$	H	0	
I-240	4-Cl		H	H	H	$COOC_2H_5$	H	1	
I-241	4-Cl		H	H	H	$COOC_2H_5$	H	2	
I-242	4-Cl		H	H	H	$COOC_2H_5$	H	0	

Table 10

Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})
I-243	4-Cl		H	H	H	$COOC_2H_5$	H	1	
I-244	4-Cl		H	H	H	$COOC_2H_5$	H	2	
I-245	4-Cl		H	H	H	$COOC_2H_5$	H	0	
I-246	4-Cl		H	H	H	$COOC_2H_5$	H	1	
I-247	4-Cl		H	H	H	$COOC_2H_5$	H	2	76- 79
I-248	4-Cl	CH_3	CH_3	CH_3	H	COC_5H_{11}	CH3	2	94- 96
I-249	H	CH_3	H	H	H	$COOC_2H_5$	H	2	143-144
I-250	H	CF_3	H	H	H	COC_2H_5	H	2	107-109
I-251	H	CF_3	H	H	H	H	H	2	103-110
I-252	4-F	CF_3	H	H	H	H	H	2	73- 76
I-253	4-F	CF_3	H	H	H	COC_2H_5	H	2	129-130
I-254	4-F	CH_3	H	H	H	COOCH3	H	0	1.6182
I-255	4-F	CH_3	H	H	H	COOCH3	H	2	167-169
I-256	4-F	CH_3	H	H	H	$COOC_2H_5$	H	2	148-149
I-257	4-F	C_2H_5	H	H	H	$COOCH_3$	H	0	84- 85
I-258	4-Br	CF_3	H	H	H	COC_2H_5	H	2	143-145
I-259	4-Br	CH_3	H	H	H	$COOC_2H_5$	H	2	158-159
I-260	4-Br	CF_3	H	H	H	H	H	2	66- 68
I-261	2-F, 4-Cl	CF_3	H	H	H	$COOC_2H_5$	H	0	83- 85
I-262	3, 4, 5-Cl_3	CF_3	H	H	H	$COOC_2H_5$	H	2	
I-263	3-F, 4-Cl	CF_3	H	H	H	$COOC_2H_5$	H	2	54- 55
I-264	3-F, 4-Cl	CH_3	H	H	H	$COOC_2H_5$	H	2	54- 56
I-265	4-CH_3	CF_3	H	H	H	COC_2H_5	H	2	115-118
I-266	4-CH_3	CH_3	H	H	H	$COOC_2H_5$	H	2	203-204

Table 11

Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})
I-267	4-C_4H_9-t	CF_3	H	H	H	$COOC_2H_5$	H	2	164-166
I-268	4-CF_3	CF_3	H	H	H	H	H	2	1.5592
I-269	4-OCH3	CF_3	H	H	H	H	H	2	135-138
I-270	4-CHF_2	CF_3	H	H	H	$COOC_2H_5$	H	2	
I-271	4-CHF_2	CF_3	H	H	H	$COOC_2H_5$	H	1	
I-272	4-CHF_2	CF_3	H	H	H	$COOC_2H_5$	H	0	
I-273	4-SO_2CH_3	CH_3	H	H	H	$COOC_2H_5$	H	2	120-122
I-274	4-CF_3	CF_3	H	H	H	COC_2H_5	H	2	189-190
I-275	4-CF_3	CH_3	H	H	H	$COOC_2H_5$	H	2	
I-276	4-CF_3	CH_3	H	H	H	$COOC_2H_5$	H	1	
I-277	4-CN	CF_3	H	H	H	COC_2H_5	H	2	195-197
I-278	4-CN	CF_3	H	H	H	H	H	2	95- 96
I-279	4-CN	CH_3	H	H	H	$COOC_2H_5$	H	2	
I-280	4-NO_2	CF_3	H	H	H	COC_2H_5	H	2	193-194
I-281	4-NO_2	CF_3	H	H	H	H	H	2	130-133
I-282	4-NO_2	CF_3	H	H	H	$COOC_2H_5$	H	2	65- 67
I-283	4-OCH_3	CF_3	H	H	H	COC_2H_5	H	2	121-123
I-284	4-OCH_3	CH_3	H	H	H	$COOC_2H_5$	H	2	151-152
I-285	4-Cl	CH_3	H	H	H	-$COCH_2CH_2CH_2$-		2	117-120
I-286	4-Cl	-$CH_2CH_2CH_2$-		H	H	$COOC_2H_5$	H	2	100-103
I-287	4-Cl	-$CH_2CH_2CH_2$-		H	H	$COOC_2H_5$	H	0	
I-288	4-Cl	-CH_2CH_2S-		H	H	$COOC_2H_5$	H	0	
I-289	4-Cl	-CH_2CH_2O-		H	H	$COOC_2H_5$	H	0	
I-290	4-Cl	CH_3	H	H	H	COC_4H_9	$CH_2OC_2H_5$	2	1.5838
I-291	4-Cl	CH_3	H	H	H	COC_3H_7	$CH_2OC_2H_5$	2	1.5835
I-292	4-Cl	CH_3	H	H	H	COC_7H_{15}	H	2	121-123
I-293	4-Cl	CH_3	H	H	H	COC_2H_5	CH_3	2	171-173

Table 12

Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})
I-294	3-F, 4-Cl	CH_3	H	H	H	$COOCH_3$	H	2	83- 85
I-295	4-Cl	CH_3	H	H	H	$COOCH_3$	C_2H_5	0	1.6046
I-296	4-Cl	CH_3	H	H	H	$COOCH_3$	C_2H_5	1	48- 50
I-297	4-Cl	CH_3	H	H	H	$COOCH_3$	C_2H_5	2	56-58
I-298	4-Cl	CH_3	H	H	H	$COOCH_3$	$CH_2OC_2H_4OCH_3$	0	1.5889
I-299	4-Cl	CH_3	H	H	H	$COOCH_3$	$CH_2OC_2H_4OCH_3$	2	1.5651
I-300	4-Cl	CH_3	H	H	H	$COOCH_3$	$CH_2OC_2H_4OCH_3$	1	1.5931
I-301	4-Cl	CH_3	H	H	H	COC_2H_5	$CH_2OC_2H_5$	0	1.6019
I-302	4-Cl	CH_3	H	H	H	COC_2H_5	$CH_2OC_2H_5$	2	41- 43
I-303	4-Cl	CH_3	H	H	H	$COCH_2OC_2H_5$	H	0	79- 80
I-304	4-Cl	CH_3	H	H	H	$COCH_2OC_2H_5$	H	2	76- 78
I-305	4-Cl	CH_3	H	H	H	$COCH_2OC_2H_5$	$CH_2OC_2H_5$	0	1.5909
I-306	4-Cl	CH_3	H	H	H	$COCH_2OC_2H_5$	$CH_2OC_2H_5$	2	1.5869
I-307	4-Cl	CH_3	H	H	H	$COOCH_3$	CH_2-  -Cl	2	65- 67
I-308	4-Cl	CH_3	H	H	H	$COOCH_3$	CH_2- 	0	1.6670
I-309	4-Cl	CH_3	H	H	H	$COOCH_3$	CH_2- 	2	59- 60
I-310	4-Cl	CH_3	H	H	H	$C(CH_3)=CCOCH_3$	H	2	182-184
I-311	4-Cl	CH_3	H	H	H	$COOCH_3$	C_4H_9	0	1.5714
I-312	4-Cl	CH_3	H	H	H	$COOCH_3$	C_4H_9	2	42- 43
I-313	4-Cl	CH_3	H	H	3-F	$COOCH_3$	H	2	109-110
I-314	4-Cl	CH_3	H	H	3-F	$COOC_2H_5$	H	2	144-146
I-315	4-Cl	CH_3	H	H	H	$COOC_5H_{11}$	H	2	59- 61
I-316	4-Cl	CH_3	H	H	H	$COOC_4H_9$	H	0	74- 75
I-317	4-F	CH_3	H	H	H	$COOCH_3$	$CH_2OC_2H_5$	0	1.5791
I-318	4-F	CH_3	H	H	H	$COOCH_3$	$CH_2OC_2H_5$	2	38- 40
I-319	4-F	CH_3	H	H	H	$COOCH_3$	$CH_2OC_2H_4OCH_3$	0	1.5732
I-320	4-F	CH_3	H	H	H	$COOCH_3$	$CH_2OC_2H_4OCH_3$	2	1.5702

Table 13

Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})
I-321	4-F	CH_3	H	H	H	$COOCH_3$	CH_2--CF_3	0	1.5699
I-322	4-$CH_2SO_2CH_3$	CH_3	H	H	H	$COOC_2H_5$	H	2	193-194
I-323	4-F	CH_3	H	H	H	$COOCH_3$	CH_2-  -CF_3	2	57- 58
I-324	4-$OCHF_2$	CH_3	H	H	H	$COOCH_3$	H	2	93- 95
I-325	4-OCH_2CF_3	CH_3	H	H	H	$COOCH_3$	H	2	152-154
I-326	4-OCF_3	CH_3	H	H	H	$COOC_2H_5$	H	2	151-153
I-327	4-OSO_2CF_3	CH_3	H	H	H	$COOC_2H_5$	H	2	163-164
I-328	4-Cl	C_2H_5	H	H	H	$COOCH_3$	$CH_2OC_2H_5$	0	1.5831
I-329	4-Cl	C_2H_5	H	H	H	$COOCH_3$	$CH_2OC_2H_5$	2	1.5735
I-330	4-Cl	C_2H_5	H	H	H	$COOCH_3$	$CH_2OC_2H_5$	1	1.5812
I-331	4-Cl	C_2H_5	H	H	H	COC_5H_{11}	CH_3	0	1.6025
I-332	4-Cl	C_2H_5	H	H	H	COC_5H_{11}	CH_3	2	1.5918
I-333	4-Cl	C_2H_5	H	H	H	COC_5H_{11}	$CH_2OC_2H_5$	0	1.5725
I-334	4-Cl	C_2H_5	H	H	H	COC_5H_{11}	$CH_2OC_2H_5$	2	1.5659
I-335	4-Cl	C_2H_5	H	H	H	$COOCH_3$	$CH_2OC_2H_4OCH_3$	2	1.5751
I-336	4-Cl	C_2H_5	H	H	H	$COOC_4H_9$	H	0	1.5899
I-337	4-Cl	C_2H_5	H	H	H	$COC_5H_{11}H$	$CH_2OC_2H_5$	1	1.5682
I-338	4-Cl	C_2H_5	H	H	H	$COOC_4H_9$	H	1	1.6029
I-339	4-Cl	C_2H_5	H	H	H	$COOC_4H_9$	$CH_2OC_2H_5$	0	1.5631
I-340	4-Cl	C_2H_5	H	H	H	$COOC_4H_9$	$CH_2OC_2H_5$	2	1.5603
I-341	4-Cl	C_2H_5	H	H	H	$COOC_4H_9$	$CH_2OC_2H_5$	1	1.5728
I-342	4-Cl	C_2H_5	H	H	H	CO-  -Cl	H	0	1.6557
I-343	2-F, 4-Cl	C_2H_5	H	H	H	$COOCH_3$	H	2	172-174
I-344	3-F, 4-Cl	C_2H_5	H	H	H	$COOCH_3$	H	2	165-167
I-345	4-Cl	C_2H_5	H	H	H	CO-  -Cl	H	1	78- 80
I-346	4-Cl	C_2H_5	H	H	H	$COCH_3$	COOCH3	0	1.6201

Table 14

Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})
I-347	4-Cl	C_2H_5	H	H	H	$COCH_3$	$COOCH_3$	1	Unmeasurable
I-348	4-Cl	C_2H_5	H	H	H	$COOCH_3$	$COOCH_3$	1	40- 42
I-349	4-Cl	C_2H_5	H	H	H	$COCH_2OCH_3$	$COOCH_3$	0	1.5963
I-350	4-Cl	C_2H_5	H	H	H	$COCH_2OCH_3$	$COOCH_3$	1	1.5891
I-351	4-Cl	C_2H_5	H	H	H	COCH2OCH3	COOCH3	2	53- 55
I-352	4-Cl	C_2H_5	H	H	H	$COOC_2H_5$	$COOCH_3$	0	1.5940
I-353	4-Cl	C_2H_5	H	H	H	$COOC_2H_5$	COOCH3	2	50- 52
I-354	3-F, 4-Cl	C_2H_5	H	H	H	$COOCH_3$	H	0	1.6150
I-355	4-Cl	C_2H_5	H	H	H	$COCH_3$	$COOCH_3$	2	Unmeasurable
I-356	3-F, 4-Cl	C_2H_5	H	H	H	$COOCH_3$	H	1	54- 56
I-357	4-F	C_2H_5	H	H	H	$COOCH_3$	H	2	153-155
I-358	3, 4-F_2	C_2H_5	H	H	H	$COOCH_3$	H	2	121-122
I-359	4-F	C_2H_5	H	H	H	$COOC_2H_5$	H	2	134-135
I-360	3, 4-F_2	C_2H_5	H	H	H	$COOCH_3$	H	1	1.5923
I-361	H	C_2H_5	H	H	H	$COOCH_3$	H	2	149-150
I-362	3-F	C_2H_5	H	H	H	$COOCH_3$	H	2	124-126
I-363	3, 4, 5-F_3	C_2H_5	H	H	H	$COOCH_3$	H	2	184-186
I-364	4-F	C_2H_5	H	H	H	$COOCH_3$	H	1	Unmeasurable
I-365	4-Cl	CF_3	H	H	H	$CON(CH_3)_2$	H	2	144-146
I-366	4-Cl	CF_3	H	H	H	$COOCH_3$	$CH_2OC_2H_5$	0	1.5601
I-367	4-Cl	CF_3	H	H	H	$COOCH_3$	$CH_2OC_2H_5$	1	1.5628
I-368	4-Cl	CF_3	H	H	H	$COOCH_3$	CH_3	2	1.3615
I-369	4-Cl	CF_3	H	H	H	$COOCH_3$	CH_2-  -CF_3	2	1.5398
I-370	2-F, 4-Cl	CF_3	H	H	H	$COOCH_3$	H	2	165-167
I-371	4-Cl	CF_3	H	H	H	$COOCH_3$	CH_2-  -Cl	2	1.5818
I-372	4-Cl	CF_3	H	H	3-F	$COOCH_3$	H	2	85- 87

Table 15

Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})
I-373	4-Cl	CF_3	H	H	H	$COOCH_3$		2	1.5852
I-374	4-Cl	CF_3	H	H	H	$COOCH_3$		2	1.5802
I-375	4-Cl	CF_3	H	H	H	$COOCH_3$		2	45- 46
I-376	3-F, 4-Cl	CF_3	H	H	H	CH_2CF_3	H	2	1.5360
I-377	3-F, 4-Cl	CF_3	H	H	H	CH2CF3	$CONH_2$	2	40- 41
I-378	3-F, 4-Cl	CF_3	H	H	H	$COOCH_3$	H	2	129-131
I-379	3-F, 4-Cl	CF_3	H	H	H	$COOCH_3$	H	0	1.5821
I-380	4-Cl	CF_3	H	H	3-F	$COOC_2H_5$	H	1	139-140
I-381	4-Cl	CF_3	H	H	H	H	H	1	1.6052
I-382	4-Cl	CF_3	H	H	H	$COCH_2C\equiv CH$	H	2	121-122
I-383	3, 4-Cl_2	CF_3	H	H	H	$COOCH_3$	H	2	163-165
I-384	3-F, 4-Cl	CF_3	H	H	H	$COOC_2H_5$	H	0	1.5720
I-385	4-Cl	CF_3	H	H	H	$COCH_2OCH_2CF_3$	H	0	1.5579
I-386	4-Cl	CF_3	H	H	H	$COCH_2OCH_2CF_3$	H	1	127-129
I-387	3-F, 4-Cl	CF_3	H	H	H	$COOCH_3$	H	1	65- 67
I-388	4-Cl	CF_3	H	H	3-F	$COOC_2H_5$	H	0	1.5747
I-389	4-Cl	CF_3	H	H	3-F	$COOCH_3$	H	0	Unmeasurable
I-390	4-Cl	CF_3	H	H	H	$COOC_2H_5$	$COOC_2H_5$	0	1.5564
I-391	4-Cl	CF_3	H	H	H	$COOC_3H_7$	H	0	1.5793
I-392	4-Cl	CF_3	H	H	H	$COOC_4H_9$	H	0	1.5712
I-393	3-CH_3, 4-Cl	CF_3	H	H	H	$COOCH_3$	H	2	149-151
I-394	4-Cl	CF_3	CH_3	H	H	$COOC_2H_5$	H	0	1.5703
I-395	4-Cl	CF_3	CH_3	H	H	$COOCH_3$	H	0	1.5770
I-396	4-Cl	CF_3	H	H	H	COC_3H_7-i	H	0	41- 42
I-397	4-Cl	CF_3	H	H	H	$COOC_2H_4OC_2H_5$	H	0	38- 39
I-398	4-Cl	CF_3	H	H	H	$COOC_3H_7$-i	H	0	1.5930

Table 16

Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p. (°C) or refractive index (n_D^{20})
I-399	4-Cl	CF_3	H	H	H	COO-Ph	H	0	1.5992
I-400	3.4-Cl_2	CF_3	H	H	H	$COOC_2H_5$	H	0	68- 70
I-401	4-Cl	CF_3	H	H	H	$COOC_3H_7$	H	2	118-120
I-402	4-Cl	CF_3	H	H	H	$COCH_2CH_2SCH_3$	H	0	74- 76
I-403	4-Cl	CF_3	H	H	H	CO-(2,6-Cl_2-Ph)	H	0	1.6058
I-404	4-Cl	CF_3	H	H	H	CO-Ph-C_4H_9-t	H	0	141-143
I-405	4-Cl	CF_3	H	H	H	CO-Ph-O-Ph	H	0	1.6289
I-406	4-Cl	CF_3	H	H	H	CO-(2-CF_3-Ph)	H	0	151-153
I-407	4-Cl	CF_3	H	H	H	CO-Ph-CH_3	H	0	136-138
I-408	4-Cl	CF_3	H	H	H	CO-Ph-OCH_3	H	0	1.6164
I-409	4-Cl	CF_3	H	H	H	CO-Ph-NO_2	H	0	153-155
I-410	4-Cl	CF_3	H	H	H	CO-(2-furyl)	H	0	136-137
I-411	4-Cl	CF_3	H	H	H	CO-(1-naphthyl)	H	0	63- 64
I-412	4-Cl	CF_3	H	H	H	$COCH=CHCH_3$	H	0	1.6004
I-413	4-Cl	CF_3	H	H	H	CO-(2,3,5,6-F_4-4-F-Ph)	H	0	163-165
I-414	4-Cl	CF_3	H	H	H	CO-(2-thienyl)	H	0	125-127
I-415	4-F	CF_3	H	H	H	$COOCH_3$	H	0	1.5732

Table 17

Comp. No.	R^9_m	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p. (°C) or refractive index (n_D^{20})
I-416	4-F	CF_3	H	H	H	$COOCH_3$	H	1	138-139
I-417	4-F	CF_3	H	H	H	$COOCH_3$	H	2	155-157
I-418	4-F	CF_3	H	H	H	$COCH_3$	H	2	117-119
I-419	4- 	CF_3	H	H	H	$COOC_2H_5$	H	2	83- 85
I-420	3, 4-F_2	CF_3	H	H	H	COOCH3	H	2	163-165
I-421	4-$OCHF_2$	CF_3	H	H	H	$COOCH_3$	H	2	130-131
I-422	4-O- 	CF_3	H	H	H	$COOC_2H_5$	H	2	66- 68
I-423	3, 4, 5-F_3	CF_3	H	H	H	$COOCH_3$	H	2	153-154
I-424	4-OCF_3	CF_3	H	H	H	$COOCH_3$	H	2	134-135
I-425	4-OCF_3	CF_3	H	H	H	$COOC_2H_5$	H	2	62- 64
I-426	4-O-  -Cl	CF_3	H	H	H	$COOC_2H_5$	H	2	59- 60
I-427	4-OSO_2CF_3	CF_3	H	H	H	$COOC_2H_5$	H	2	61- 63
I-428	4-OCH_2CF_3	CF_3	H	H	H	$COOCH_3$	H	2	163-166
I-429	4-SCH_3	CF_3	H	H	H	$COOCH_3$	H	2	120-122
I-430	4-$SCHF_2$	CF_3	H	H	H	$COOCH_3$	H	2	139-140
I-431	3, 5-F_2	CF_3	H	H	H	$COOCH_3$	H	2	141-142
I-432	3-F	CF_3	H	H	H	$COOCH_3$	H	2	141-143
I-433	3-F	CF_3	H	H	H	CONH-  -OCF_3	H	2	108-110
I-434	3-F	CF_3	H	H	H	CONH--Cl	H	2	172-173
I-435	3-Cl, 4-F	CF_3	H	H	H	$COOCH_3$	H	2	186-188
I-436	H	CF_3	H	H	H	$COOCH_3$	H	0	1.5839
I-437	4-Br	CF_3	H	H	H	$COOCH_3$	H	0	1.5940
I-438	H	CF_3	H	H	H	$COOC_2H_5$	H	0	1.5760
I-439	4-Br	CF_3	H	H	H	$COOC_2H_5$	H	0	1.5901
I-440	H	CF_3	H	H	H	$COOCH_3$	H	1	135-136

Table 18

Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})
I-441	4-Br	CF_3	H	H	H	$COOCH_3$	H	1	169-170
I-442	4-CF_3	CF_3	H	H	H	$COOC_2H_5$	H	0	52- 54
I-443	3, 4-(OCH_2O)	CF_3	H	H	H	$COOCH_3$		2	182-185
I-444	H	CF_3	CH_3	H	H	$COOC_2H_5$	H	0	1.5680
I-445	4-F	CF_3	H	H	H	$COOC_2H_5$	H	0	1.5610
I-446	4-OCH_3	CF_3	H	H	H	$COOC_2H_5$	H	0	1.5812
I-447	4-Br	CF_3	CH_3	H	H	$COOCH_3$	H	0	1.5921
I-448	4-Br	CF_3	H	H	H	$COOC_2H_5$	H	0	1.5869
I-449	4-I	CF_3	H	H	H	$COOCH_3$	H	0	Unmeasurable
I-450	4-I	CF_3	H	H	H	$COOC_2H_5$	H	0	1.6060
I-451	4-Cl	CHF_2	H	H	H	$COOCH_3$	CH_3	0	1.6131
I-452	4-Cl	CHF_2	H	H	H	$COOCH_3$	$CH_2OC_2H_5$	0	1.5738
I-453	4-Cl	CHF_2	H	H	H	$COOCH_3$	CH_3	1	1.5991
I-454	4-Cl	CHF_2	H	H	H	$COOCH_3$	$CH_2OC_2H_5$	1	1.5802
I-455	4-Cl	CHF_2	H	H	H	$COOCH_3$	CH_2--Cl	0	Unmeasurable
I-456	4-Cl	CHF_2	H	H	H	$COOCH_3$	CH_2-  -Cl	1	Unmeasurable
I-457	4-Cl	CHF_2	H	H	H	$COOCH_3$	CH_2--CF_3	0	1.5719
I-458	4-Cl	CHF_2	H	H	H	$COOCH_3$	CH_2-  -Cl	2	Unmeasurable
I-459	4-Cl	CHF_2	H	H	H	$COOCH_3$	CH_2-  -CF_3	2	1.5500
I-460	4-Cl	CHF_2	H	H	H	$COOCH_3$	Cl CH_2-	0	1.5882
I-461	4-Cl	CHF_2	H	H	H	$COOCH_3$	Cl CH_2- 	2	47- 48
I-462	4-Cl	CHF_2	CH_3	H	H	$COOCH_3$	CH_2- 	2	1.5818

Table 19

Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})
I-463	4-Cl	CHF_2	H	H	H	$COOC_2H_5$	CH_2-Ph-Cl	2	Unmeasurable
I-464	4-Cl	CHF_2	H	H	H	$COOCH_3$	CH_2-Ph(Cl)	2	1.5950
I-465	4-Cl	CHF_2	H	H	H	$COOCH_3$	CH_2-Ph-CH_3	2	56- 57
I-466	4-Cl	CHF_2	H	H	H	$COOCH_3$	CH_3	2	1.5825
I-467	4-Cl	CHF_2	H	H	H	$COOC_2H_5$	CH3	2	1.5659
I-468	4-Cl	CHF_2	H	H	H	$COOCH_3$	C_2H_5	2	1.5775
I-469	4-Cl	CHF_2	H	H	H	$COOCH_3$	C_4H_9	2	1.5682
I-470	4-Cl	CHF_2	H	H	H	H	H	1	1.6203
I-471	3-F, 4-Cl	CHF_2	H	H	H	$COOCH_3$	H	0	111-113
I-472	4-Cl	CHF_2	H	H	H	COCH3	H	1	135-137
I-473	4-Cl	CHF_2	H	H	H	$COOC_2H_5$	H	0	1.6061
I-474	4-Cl	CHF_2	H	H	H	$COCH_2OCH_2CF_3$	H	0	1.5761
I-475	4-Cl	CHF_2	H	H	H	$COCH_2OCH_2CF_3$	H	1	1.5742
I-476	4-Cl	CHF_2	H	H	H	$COOC_2H_5$	H	1	189-190
I-477	4-Cl	CHF_2	H	H	H	CONH-Ph(Cl,Cl)	H	0	124-126
I-478	4-Cl	CHF_2	H	H	H	CONH-Ph(Cl,Cl)	H	1	102-103
I-479	4-Cl	CHF_2	H	H	H	CONH-Ph-OCF_3	H	0	90- 92
I-480	4-Cl	CHF_2	H	H	H	$COOC_3H_7$	H	0	1.6019
I-481	4-Cl	CHF_2	H	H	H	$COOCH_3$	$COOC_2H_5$	0	1.5502
I-482	3-F, 4-Cl	CHF_2	H	H	H	$COOC_2H_5$	H	0	1.5981
I-483	3-F, 4-Cl	CHF_2	H	H	H	$COOC_2H_5$	$COCH_2OCH_3$	0	1.5688
I-484	4-Cl	CHF_2	CH_3	H	H	$COOC_2H_5$	H	0	1.5912

Table 20

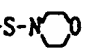
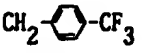
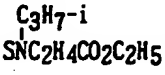
Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})
I-485	4-Cl	CHF_2	H	H	H	$COCH_3$	H	0	109-112
I-486	4-Cl	CHF_2	H	H	H	COC_3H_7	H	0	1.6049
I-487	4-Cl	CHF_2	H	H	H	$COCH_3$	H	2	196-198
I-488	4-Cl	CHF_2	H	H	H	COC_3H_7	H	2	112-114
I-489	4-Cl	CHF_2	H	H	H	$COOC_3H_7$	H	2	1.5725
I-490	4-F	CHF_2	H	H	H	$COOCH_3$	H	0	1.5947
I-491	4-F	CHF_2	H	H	H	$COOCH_3$	H	1	135-137
I-492	4-F	CHF_2	H	H	H	$COOCH_3$	H	2	148-150
I-493	4-F	CHF_2	H	H	H	$COOC_2H_5$	H	0	1.5870
I-494	4-Br	CHF_2	H	H	H	$COOCH_3$	H	0	1.6139
I-495	4-Br	CHF_2	H	H	H	$COOCH_3$	H	1	88- 90
I-496	4-Br	CHF_2	H	H	H	$COOC_2H_5$	H	0	1.6100
I-497	3.4-F_2	CHF_2	H	H	H	$COOCH_3$	H	0	133-134
I-498	3.4-F_2	CHF_2	H	H	H	$COOCH_3$	H	1	127-128
I-499	4-CF_3	CHF_2	H	H	H	$COOC_2H_5$	H	0	1.5662
I-500	4-CF_3	CHF_2	H	H	H	$COOC_2H_5$	H	1	70- 72
I-501	4-CF_3	CHF_2	H	H	H	$COOCH_3$	H	1	64- 66
I-502	4-Cl	CH_2Cl	H	H	H	$COOCH_3$	CH_3	2	1.6059
I-503	4-Cl	CF_3	H	H	H	$COOC_2H_5$		0	
I-504	4-Cl	CH_2Cl	H	H	H	$COOCH_3$		2	1.5631
I-505	4-Cl	CF_3	H	H	H	$COCH_3$		0	
I-506	4-Cl		H	H	H	$COOC_2H_5$	H	2	134-135
I-507	4-Cl	$CONHC_4H_9$	H	H	H	$COOCH_3$	H	0	47- 49
I-508	4-Cl	CF_3	H	H	H	$COOCH_3$	$SN(C_4H_9)_2$	0	

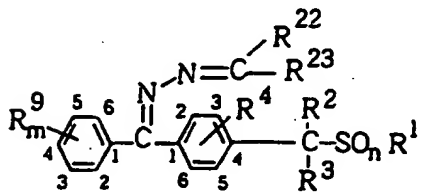
Table 21

Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})
I-509	4-Cl	CH_2Cl	H	H	H	CO-(pyridyl)-Cl	H	2	132-135
I-510	4-F	CH_2Cl	H	H	H	$COOCH_3$	H	2	152-154
I-511	4-Cl	$N(CH_3)_2$	H	H	H	$COOCH_3$	H	2	
I-512	4-Cl	-N(pyrrolidine)	H	H	H	$COCH_3$	H	2	
I-513	4-Cl	NHC_2H_5	H	H	H	$COOC_2H_5$	H	2	
I-514	4-Cl	-N=C(NH-CH_3)-S	H	H	H	$COOC_2H_5$	H	2	
I-515	4-Cl	-(triazole)	H	H	H	$COOC_2H_5$	H	2	
I-516	4-Cl	C_2H_5	H	H	H	$-C(C_2H_5)=NSO_2$-(phenyl)	H	0	1.6408
I-517	4-Cl	C_2H_5	H	H	H	$-C(C_2H_5)=NSO_2$-(phenyl)	H	2	48- 50
I-518	4-Cl	C_2H_5	H	H	H	$-C(C_2H_5)=NSO_2$-(phenyl)-Cl	H	0	1.6424
I-519	4-Cl	C_2H_5	H	H	H	$-C(C_2H_5)=NSO_2$-(phenyl)-Cl	H	2	75- 76
I-520	4-Cl	CF_3	$COOC_2H_5$	$COOC_2H_5$	H	$COOC_2H_5$	H	0	1.5345
I-521	4-Br	C_2H_5	H	H	H	$COOCH_3$	H	2	172-174
I-522	4-Br	C_2H_5	H	H	H	$COOC_2H_5$	H	2	96- 98
I-523	4-Br	C_2H_5	H	H	H	H	H	2	1.6258
I-524	4-Br	C_2H_5	H	H	H	$COCH_3$	H	2	202-204
I-525	4-Br	C_2H_5	H	H	H	$COOC_3H_7$	H	2	105-108
I-526	4-Br	C_2H_5	H	H	H	$COOC_2H_4OC_2H_5$	H	2	65- 67
I-527	4-Br	C_2H_5	H	H	H	H	H	0	1.6555
I-528	4-Br	C_2H_5	H	H	H	$COOCH_3$	H	0	1.6415

Table 22

Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	R^{12}	R^{13}	n	m.p. (°C) or refractive index (n_D^{20})
I-529	4-Br	C_2H_5	H	H	H	$COOC_2H_5$	H	0	1.6221
I-530	4-Br	C_2H_5	H	H	H	$COCH_3$	H	0	1.6396
I-531	4-Br	C_2H_5	H	H	H	COC_2H_5	H	0	1.6463
I-532	4-Br	C_2H_5	H	H	H	$COOCH_3$	H	1	70- 72
I-533	4-Br	C_2H_5	H	H	H	$COOC_2H_5$	H	1	62- 64
I-534	4-Br	C_2H_5	H	H	H	COC_2H_5	H	1	60- 62
I-535	4-Br	CHF_2	H	H	H	$COCH_3$	H	0	105-108
I-536	4-Br	CHF_2	H	H	H	COC_2H_5	H	0	1.6328
I-537	4-Br	CHF_2	H	H	H	$COCH_3$	H	1	138-139
I-538	4-Br	CHF_2	H	H	H	COC_2H_5	H	1	135-137
I-539	4-Br	C_2H_5	H	H	H	$COOCH_3$	CH_2CN	2	74- 76
I-540	4-Cl	CF_3	H	H	H	$COOC_2H_5$	CH_2CN	0	1.5689

Table 23



Comp. No.	R^9_m	R^1	R^2	R^3	R^4	R^{22}	R^{23}	n	m.p.(°C) or refractive index (n_D^{20})
II-1	4-Cl	CH_3	H	H	H	H		2	
II-2	4-Cl	CH_3	H	H	H	H	$N(CH_3)OCH_3$	1	
II-3	4-Cl	CH_3	H	H	H	H	$N(CH_3)OCH_3$	2	
II-4	4-Cl	CH_3	H	H	H	Cl	$N(CH_3)OCH_3$	2	
II-5	4-Cl	CF_3	H	H	H	CH_3	$NH(CH_2)_3OCH_3$	2	1.5951
II-6	4-Cl	CF_3	H	H	H	CH_3	$N(CH_3)_2$	2	50- 52
II-7	4-Cl	CF_3	H	H	H	CH_3	$N(CH_3)OCH_3$	2	
II-8	4-Cl	CF_3	H	H	H	CH_3	$NHCH_3$	2	58- 60
II-9	4-Cl	CF_3	H	H	H	C_2H_5		0	
II-10	4-Cl	CF_3	H	H	H	C_2H_5		2	1.5978
II-11	4-Cl	CF_3	H	H	H	C_2H_5	$N(CH_3)_2$	2	51-53
II-12	4-Cl	CF_3	H	H	H	C_2H_5	$N(CH_3)_2$	0	1.6238
II-13	4-Cl	CF_3	H	H	H	C_2H_5	$NHCH_3$	2	55- 57
II-14	4-Cl	CF_3	H	H	H	C_2H_5	Cl	2	108-109
II-15	4-Cl	CF_3	H	H	H	C_4H_9	$N(CH_3)_2$	2	1.6049
II-16	4-Cl	CF_3	H	H	H	C_4H_9	$NHCH_3$	2	35- 38
II-17	4-Cl	CH_3	H	H	H	CH_3		2	
II-18	4-Cl	CH_3	H	H	H	CH_3	C_4H_9	2	
II-19	4-Cl	CH_3	H	H	H	CH_3	C_4H_9	1	
II-20	4-Cl	CH_3	H	H	H	CH_3	CH_3	2	
II-21	4-Cl	CH_3	H	H	H	CH_3	$N(C_4H_9\text{-}t)OCH_3$	2	

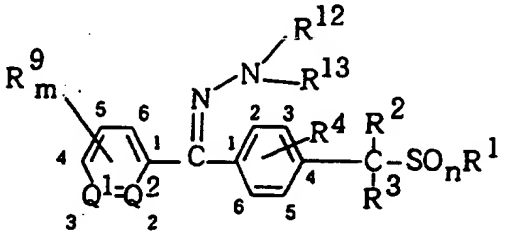
Table 24

Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	R^{22}	R^{23}	n	m.p.(°C) or refractive index (n_D^{20})
II-22	4-Cl	CH_3	H	H	H	CH_3	$N(CH_3)_2$	0	Unmeasurable
II-23	4-Cl	CH_3	H	H	H	CH_3	$N(CH_3)_2$	1	53- 55
II-24	4-Cl	CH_3	H	H	H	CH_3	$N(CH_3)_2$	2	156-158
II-25	4-Cl	CH_3	H	H	H	CH_3	$N(CH_3)OC_4H_9$-t	2	
II-26	4-Cl	CH_3	H	H	H	CH_3	$N(CH_3)OCH_3$	2	
II-27	4-Cl	CH_3	H	H	H	CH_3	NH_2	2	
II-28	4-Cl	CH_3	H	H	H	CH_3	NH_2	1	
II-29	4-Cl	CH_3	H	H	H	C_2H_5		0	1.6498
II-30	4-Cl	CH_3	H	H	H	C_2H_5		1	Unmeasurable
II-31	4-Cl	CH_3	H	H	H	C_4H_9	$N(CH_3)OCH_3$	2	
II-32	4-Cl	CH_3	H	H	H	C_6H_{13}	$N(CH_3)_2$	2	115-118
II-33	4-Cl	CH_3	H	H	H	C_6H_{13}	$N(CH_3)_2$	1	
II-34	4-Cl	CH_3	H	H	H	C_6H_{13}	$N(CH_3)_2$	0	
II-35	4-Cl	C_2H_5	H	H	H	CH_3	CH_3	2	1.6163
II-36	4-Cl	CH_2Cl	H	H	H	C_2H_5	Cl	2	1.6108
II-37	4-Cl	CHF_2	CH_3	CH_3	H	H	H	0	1.6089
II-38	4-Cl	CF_3	H	H	H	H	$N(CH_3)_2$	2	1.6180
II-39	4-Cl	CF_3	H	H	H	CH_3	$N(CH_3)_2$	0	1.6248
II-40	4-Cl	CF_3	H	H	H	CH_3	$N(CH_3)_2$	1	Unmeasurable
II-41	3-F, 4-Cl	CF_3	H	H	H	CH_3	$N(CH_3)_2$	2	1.6171
II-42	4-Cl	CF_3	H	H	3-F	CH_3	$N(CH_3)_2$	2	62- 68
II-43	4-F	CF_3	H	H	H	CH_3	$N(CH_3)_2$	2	109-111
II-44	4-Br	CF_3	H	H	H	CH_3	$N(CH_3)_2$	2	123-124
II-45	4-Br	CF_3	H	H	H	C_2H_5	Cl	2	1.6207
II-46	4-Br	CF_3	H	H	H	C_2H_5	$N(CH_3)_2$	2	1.5619
II-47	4-F	CF_3	H	H	H	C_2H_5	Cl	2	1.5892
II-48	4-F	CF_3	H	H	H	C_2H_5	$N(CH_3)_2$	2	103-104

Table 25

Comp. No.	R^9_m	R^1	R^2	R^3	R^4	R^{22}	R^{23}	n	m.p.(°C) or refractive index (n_D^{20})
II-49	4-Cl	CHF_2	H	H	H	CH_3	$N(CH_3)_2$	2	1.6335
II-50	4-Cl	CHF_2	H	H	H	CH_3	$N(CH_3)_2$	1	75- 77
II-51	4-Cl	C_2H_5	H	H	H	H	$N(CH_3)_2$	2	130-132
II-52	4-Cl	CHF_2	H	H	H	C_2H_5	$N(CH_3)_2$	0	1.6479

Table 26



Comp. No.	R^9_m	R^1	R^2	R^3	R^4	R^{12}	R^{13}	Q^1	Q^2	n	m.p.(°C) or refractive index (n_D^{20})
III-1	4-Cl	CH_3	H	H	H	$COOC_2H_5$	H	N	CH	2	149-150
III-2	4-Cl	CF_3	H	H	H	$COOC_2H_5$	H	N	CH	2	72- 73
III-3	4-Cl	CF_3	H	H	H	H	H	N	CH	2	103-105
III-4	4-Cl	CF_3	H	H	H	$COCH_3$	H	N	CH	2	165-167
III-5	4-Cl	CF_3	H	H	H	$COOCH_3$	H	N	CH	2	85- 87
III-6	4-Cl	CH_2Cl	H	H	H	$COOC_2H_5$	H	N	CH	2	67- 69
III-7	4-Cl	CH_2Cl	H	H	H	$COOCH_3$	H	N	CH	2	85- 87
III-8	4-Cl	C_2H_5	H	H	H	$COOCH_3$	H	N	CH	2	176-178
III-9	4-Cl	C_2H_5	H	H	H	$COOC_2H_5$	H	N	CH	2	Unmeasurable
III-10	4-Cl	CF_3	H	H	H	$COOCH_3$	H	N	CH	0	Unmeasurable
III-11	4-Cl	CF_3	H	H	H	$COOCH_3$	H	N	CH	1	70- 71
III-12	4-Cl	CF_3	H	H	H	$COOCH_3$	H	CH	N	2	66- 67
III-13	4-Cl	CF_3	H	H	H	$COOC_2H_5$	H	CH	N	2	41- 42

Table 27

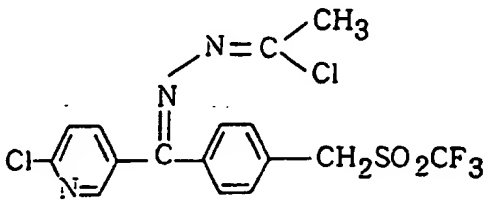
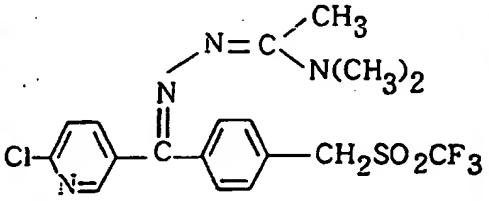
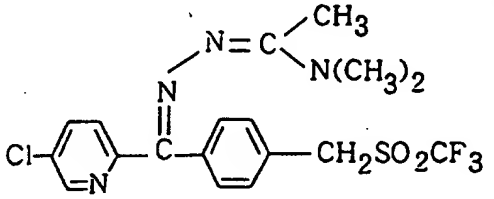
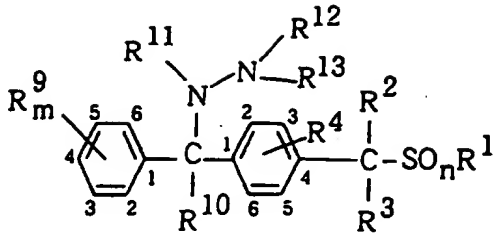
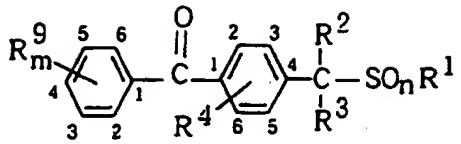
Comp. No.	Structure	m.p.(°C) or refractive index (n_D^{20})
IV - 1		1.5979
IV - 2		1.6065
IV - 3		Unmeasurable

Table 28



Comp. No.	R^9_m	R^1	R^2	R^3	R^4	R^{10}	R^{11}	R^{12}	R^{13}	n	m.p.(°C) or refractive index (n_D^{20})	Remarks
V- 1	4-Cl	CH_3	H	H	H	H	H	COC_2H_5	H	2	83- 85	
V- 2	4-Cl	C_2H_5	H	H	H	H	H	H	H	2	1.6079	
V- 3	4-Cl	C_2H_5	H	H	H	H	H	COC_2H_5	H	2	63- 64	
V- 4	4-Cl	C_2H_5	H	H	H	H	H	$COOCH_3$	H	2	60- 61	
V- 5	4-Cl	C_2H_5	H	H	H	H	H	$COOCH_3$	H	2	76- 78	Hydrochloride
V- 6	4-Cl	C_2H_5	H	H	H	H	$COOC_2H_5$	$COOC_2H_5$	H	2	66- 67	
V- 7	4-Cl	C_2H_5	H	H	H	H	$COOCH_3$	COOCH3	H	2	69- 71	
V- 8	4-Cl	CF_3	H	H	H	H	H	H	H	2	1.5671	
V- 9	4-Cl	CF_3	H	H	H	H	H	COC_2H_5	H	2	49- 50	
V-10	4-Cl	CF_3	H	H	H	H	H	$COCH_3$	H	2	49- 50	
V-11	4-Cl	CF_3	H	H	H	H	H	$COOCH_3$	H	2	47- 49	
V-12	4-Cl	CF_3	H	H	H	H	H	$COOCH_3$	H	2	1.5098	Methane sulfate
V-13	4-Cl	CF_3	H	H	H	H	H	$COOCH_3$	H	2	52- 54	Hydrochloride
V-14	4-Cl	CF_3	H	H	H	H	H	$COOC_2H_5$	H	2	Unmeasurable	
V-15	4-Cl	CF_3	H	H	H	H	$COOCH_3$	$COOCH_3$	H	2	55- 56	
V-16	4-Cl	CHF_2	H	H	H	H	H	$COOCH_3$	H	2	50- 53	
V-17	4-Cl	CHF_2	H	H	H	H	H	$COOCH_3$	H	2	87- 88	Hydrochloride
V-18	4-F	CF_3	H	H	H	H	H	H	H	2	1.5412	
V-19	4-F	CF_3	H	H	H	H	H	COC_2H_5	H	2	50- 52	

Table 29



Comp. No.	R^9_m	R^1	R^2	R^3	R^4	n	m.p.(°C) or refractive index (n_D^{20})
VI- 1	4-Cl	CH_3	H	H	H	0	59- 61
VI- 2	4-Cl	CH_3	H	H	H	1	116-118
VI- 3	4-Cl	CH_3	H	H	H	2	164-166
VI- 4	4-Cl	C_2H_5	H	H	H	0	33- 34
VI- 5	4-Cl	C_2H_5	H	H	H	1	
VI- 6	4-Cl	C_2H_5	H	H	H	2	117-118
VI- 7	4-Cl	C_3H_7	H	H	H	2	128-129
VI- 8	4-Cl	C_3H_7-i	H	H	H	2	135-137
VI- 9	4-Cl	C_4H_9	H	H	H	2	118-119
VI-10	4-Cl	CH_2Cl	H	H	H	2	148-150
VI-11	4-Cl	C_3H_6Br	H	H	H	2	105-107
VI-12	4-Cl	CF_3	H	H	H	0	63- 65
VI-13	4-Cl	CF_3	H	H	H	1	114-115
VI-14	4-Cl	CF_3	H	H	H	2	123-125
VI-15	4-Cl	CHF_2	H	H	H	0	34- 35
VI-16	4-Cl	CHF_2	H	H	H	2	154-157
VI-17	4-Cl	C_2F_5	H	H	H	0	52- 53
VI-18	4-Cl	C_2F_5	H	H	H	2	94- 96
VI-19	4-Cl	CH_2CF_3	H	H	H	2	148-150
VI-20	4-Cl	CF_2CHF_2	H	H	H	0	48- 50
VI-21	4-Cl	CF_2CHF_2	H	H	H	2	68- 70
VI-22	4-Cl	CF_2CHFCF_3	H	H	H	0	Unmeasurable
VI-23	4-Cl	CH_2CN	H	H	H	0	54- 55
VI-24	4-Cl	CH_2CN	H	H	H	2	179-181

Table 30

Comp. No.	R^9_m	R^1	R^2	R^3	R^4	n	m.p.(°C) or refractive index (n_D^{20})
VI-25	4-Cl		H	H	H	2	113-115
VI-26	4-Cl	CH_2CH_2OH	H	H	H	0	61- 62
VI-27	4-CF_3	CF_3	H	H	H	2	128-131
VI-28	4-Cl	CF_3	CH_3	CH_3	H	2	107-109
VI-29	4-Cl	CH_2CH_2OH	H	H	H	2	161-162
VI-30	4-F	CH_3	H	H	H	0	1.6141
VI-31	4-F	CH_3	H	H	H	2	138-139
VI-32	4-F	CF_3	H	H	H	0	43- 45
VI-33	4-F	C_2H_5	H	H	H	0	1.6022
VI-34	4-F	C_2H_5	H	H	H	2	97- 98
VI-35	4-Cl	CN	H	H	H	0	129-131
VI-36	4-Cl	$-CH_2CH_2CH_2-$		H	H	2	Unmeasurable
VI-37	4-Cl		H	H	H	0	80- 90
VI-38	4-Cl		H	H	H	2	101-103
VI-39	4-Cl	CH_3	H	H	3-F	2	132-133
VI-40	4-Cl	CF_3	H	H	H	2	144-145
VI-41	4-Cl	CHF_2	H	H	H	1	124-125
VI-42	4-Cl	$CONHC_4H_9$	H	H	H	0	115-116
VI-43	4-Cl	CF_3	H	H	3-F	0	1.5684
VI-44	4-Cl	CN	CH_3	H	H	0	1.6191
VI-45	4-Cl	CF_3	CH_3	H	H	0	1.5698
VI-46	4-Cl	CH_3	CH_3	H	H	2	145-147
VI-47	H	CN	H	H	H	0	138-139
VI-48	H	CN	CH_3	H	H	0	1.6189
VI-49	H	CF_3	H	H	H	0	43- 44
VI-50	H	CF_3	CH_3	H	H	0	1.5581

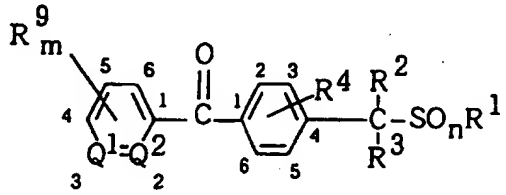
Table 31

Comp. No.	$R^9{}_m$	R^1	R^2	R^3	R^4	n	m.p.(°C) or refractive index (n_D^{20})
VI-51	H	CH_3	CH_3	H	H	0	143-145
VI-52	4-F	CF_3	H	H	H	2	84- 85
VI-53	4-F	CH_2Cl	H	H	H	2	110-112
VI-54	4-F	CHF_2	H	H	H	0	1.5572
VI-55	4-F	CHF_2	H	H	H	2	165-167
VI-56	4-F	CN	H	H	H	0	116-117
VI-57	4-$OCHF_2$	CH_3	H	H	H	2	130-131
VI-58	4-$OCHF_2$	CF_3	H	H	H	2	95- 96
VI-59	4-OCH_2CF_3	CH_3	H	H	H	2	110-112
VI-60	4-OCH_2CF_3	CF_3	H	H	H	2	105-107
VI-61	3,4,5-F_3	CF_3	H	H	H	2	83- 84
VI-62	4-OCF_3	CF_3	H	H	H	2	116-117
VI-63	3-F,4-Cl	CF_3	H	H	H	2	79- 81
VI-64	3-F,4-Cl	CN	H	H	H	0	93- 95
VI-65	3-F,4-Cl	C_2H_5	H	H	H	0	1.6129
VI-66	3-F,4-Cl	CHF_2	H	H	H	0	1.5922
VI-67	3-F,4-Cl	CF_3	H	H	H	0	69- 70
VI-68	4-Br	CN	H	H	H	0	145-148
VI-69	4-Br	CF_3	H	H	H	0	74- 75
VI-70	4-Br	CHF_2	H	H	H	0	63- 64
VI-71	4-Br	CHF_2	H	H	H	1	118-119
VI-72	4-Br	CN	CH_3	H	H	0	53- 54
VI-73	4-Br	CF_3	CH_3	H	H	0	39- 40
VI-74	3,4-F_2	C_2H_5	H	H	H	1	176-178
VI-75	4-O-  -Cl	CF_3	H	H	H	2	125-126
VI-76	4-OCH_3	CN	H	H	H	0	79- 81
VI-77	4-OCH_3	CF_3	H	H	H	0	57- 59

Table 32

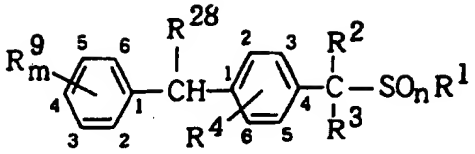
Comp. No.	R^9_m	R^1	R^2	R^3	R^4	n	m.p.(°C) or refractive index (n_D^{20})
VI-78	4-I	CN	H	H	H	0	116-118
VI-79	4-I	CF_3	H	H	H	0	92- 94
VI-80	4-Cl	CF_3	$COOC_2H_5$	$COOC_2H_5$	H	0	1.5362
VI-81	4-Cl	CCl_3	H	H	H	0	103-105
VI-82	4-Br	C_2H_5	H	H	H	2	141-142
VI-83	4-Br	CHF_2	H	H	H	2	113-115
VI-84	4-Br	C_2H_5	H	H	H	0	30
VI-85	4-Cl	CH_3	$COOC_2H_5$	$COOC_2H_5$	H	0	1.5674

Table 33



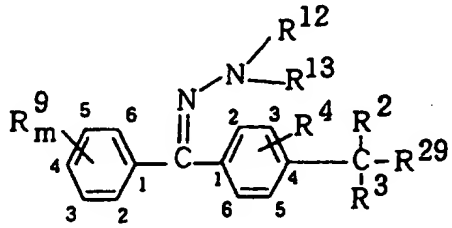
Comp. No.	R^9_m	R^1	R^2	R^3	R^4	Q^1	Q^2	n	m.p.(°C) or refractive index (n_D^{20})
VII-1	4-Cl	CH_3	H	H	H	N	CH	2	162-164
VII-2	4-Cl	CF_3	H	H	H	N	CH	2	97- 99
VII-3	4-Cl	CF_3	H	H	H	N	CH	0	1.5820
VII-4	4-Cl	CH_2Cl	H	H	H	N	CH	2	94- 96
VII-5	4-Cl	C_2H_5	H	H	H	N	CH	2	155-156
VII-6	4-Cl	CN	H	H	H	N	CH	0	93- 94
VII-7	4-Cl	CH_3	H	H	H	CH	N	2	153-155
VII-8	4-Cl	CF_3	H	H	H	CH	N	2	38- 39

Table 34



Comp. No.	R^9_m	R^1	R^2	R^3	R^4	R^{28}	n	m.p.(°C) or refractive index (n_D^{20})
VIII-1	4-Cl	C_2H_5	H	H	H	OH	2	1.5950
VIII-2	4-Cl	CF_3	H	H	H	OH	2	113-115
VIII-3	4-Cl	CF_3	H	H	H	OH	0	1.5601
VIII-4	4-Cl	CHF_2	H	H	H	OH	2	85- 87
VIII-5	4-Cl	C_2H_5	H	H	H	Cl	2	1.6044
VIII-6	4-F	CH_3	H	H	H	OH	2	139-140
VIII-7	4-F	CF_3	H	H	H	OH	2	99-100

Table 35

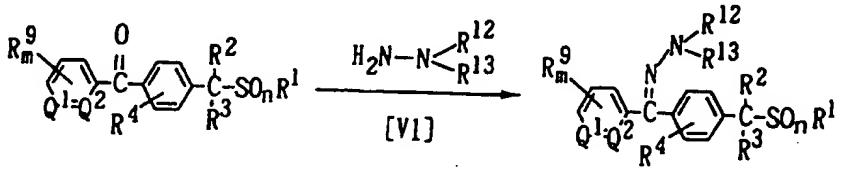


Comp. No.	R^9_m	R^2	R^3	R^4	R^{12}	R^{13}	R^{29}	m.p.(°C) or refractive index (n_D^{20})
IX-1	4-Cl	H	H	H	$COOCH_3$	H	OH	Unmeasurable
IX-2	4-Cl	H	H	H	$COOC_2H_5$	H	Cl	113-115
IX-3	4-Cl	H	H	H	$COOCH_3$	H	SH	162-165

Now, processes for producing the compounds of the present invention will be described.

The compounds of the formula (I) of the present invention can be produced in accordance with the following processes 1 to 5.

Process 1



[IV] [VI] [I]

In the above formulas, R^1, R^2, R^3, R^4, R^9, R^{12}, R^{13}, m, n, Q^1 and Q^2 are as defined above.

In Process 1, a compound of the formula (I) of the present invention wherein A is (A2) can be obtained by reacting 1 mol of a benzophenone of the formula (IV) with from 1.0 to 10.0 mols of a hydrazine of the formula (VI) or its hydrate in the presence of from 0 to 5 ℓ of a solvent, if necessary in the presence of from 0.01 to 1.0 mol of an acid catalyst.

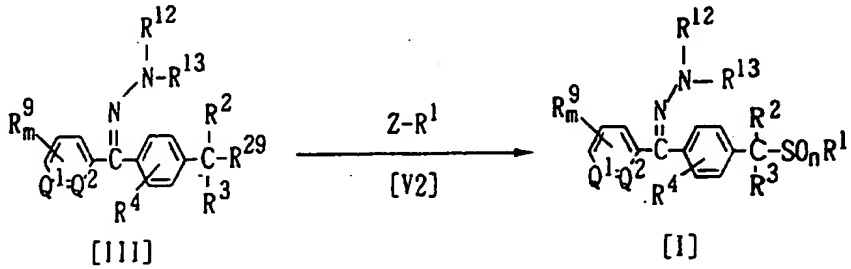
The solvent which can be used, may, for example, be an aromatic hydrocarbon such as benzene, toluene, xylene or chlorobenzene, an aprotic polar solvent such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methyl-2-pyrrolidone, dimethylsulfoxide or sulfolane, an alcohol such as methanol, ethanol, ethylene glycol or glycerol, a halogenated hydrocarbon such as methylene chloride or

chloroform, an ester such as ethyl acetate or ethyl propionate, an aliphatic hydrocarbon such as hexane, cyclohexane or heptane, a pyridine such as pyridine or picoline, acetic acid or water, or a solvent mixture thereof.

The acid catalyst may, for example, be a mineral acid such as hydrochloric acid, sulfuric acid or nitric acid, an organic acid such as formic acid, acetic acid, propionic acid, methanesulfonic acid, benzenesulfonic acid or p-toluenesulfonic acid monohydrate, an acid addition salt of an amine such as pyridine hydrochloride or triethylamine hydrochloride, a metal halide such as titanium tetrachloride, zinc chloride, ferrous chloride or ferric chloride, or boron trifluoride·etherate.

The reaction temperature is an optional temperature within a range of from -10°C to the reflux temperature in the reaction system, preferably from room temperature to 150°C. The reaction time varies depending upon the particular compound, but can be set within a range of from 10 minutes to 20 hours.

Process 2



[III] [I]

In the above formulas, R^1, R^2, R^3, R^4, R^9, R^{12}, R^{13},

m, Q^1 and Q^2 are as defined above; R^{29} is a halogen atom; Z is a group of the formula $MS(O)_n$; M is an alkali metal; and n is 0 or 2.

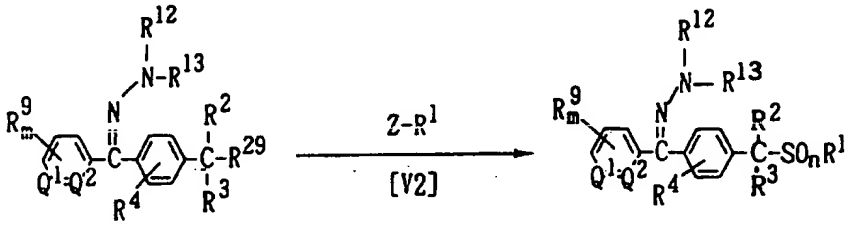
In Process 2, a compound of the formula (I) of the present invention wherein A is (A2), can be obtained by reacting 1 mol of benzyl halide of the formula (III) with from 1.0 to 3.0 mol of an alkali metal salt of a sulfur compound of the formula (V2) in the presence of from 0 to 10 ℓ of a solvent.

The solvent which can be used, may, for example, be an ether such as diethyl ether, tetrahydrofuran or dioxane, an aromatic hydrocarbon such as benzene, toluene, xylene or chlorobenzene, an aprotic polar solvent such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methyl-2-pyrrolidone, dimethylsulfoxide or sulfolane, an alcohol such as methanol, ethanol, ethylene glycol or glycerol, a halogenated hydrocarbon such as methylene chloride or chloroform, an ester such as ethyl acetate or ethyl propionate, an aliphatic hydrocarbon such as pentane, hexane, cyclohexane or heptane, a pyridine such as pyridine or picoline or water, or a solvent mixture thereof.

The alkali metal salt of the sulfur compound to be used in this process can be prepared from a sulfur compound wherein Z is $HS(O)_n$ and an alkali metal, an alkali metal hydride or an alkali metal hydroxide.

The reaction temperature is an optional temperature within a range of from -10°C to the reflux temperature in the reaction system, preferably from room temperature to 100°C. The reaction time varies depending upon the particular compound, but can be set within a range of from 10 minutes to 20 hours.

Process 3



[III] [I]

In the above formulas, R^1, R^2, R^3, R^4, R^9, R^{12}, R^{13}, m, Q^1 and Q^2 are as defined above; R^{29} is a mercapto group; and Z is a halogen atom, a C_{1-4} alkylsulfonyloxy group or a benzenesulfonyloxy group (which may be substituted by a methyl group).

In Process 3, a compound of the formula (I) of the present invention wherein A is (A2), can be obtained by reacting 1 mol of a mercapto compound of the formula (III) with from 1.0 to 5.0 mols of the compound of the formula (V2) in from 0 to 5 ℓ of a solvent in the presence of from 1.0 to 3.0 mols of a base.

The solvent which can be used, may, for example, be an ether such as diethyl ether, tetrahydrofuran or dioxane, an aromatic hydrocarbon such as benzene, toluene, xylene or chlorobenzene, an aprotic polar

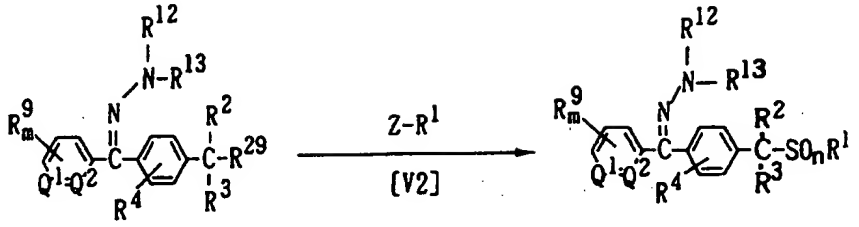
solvent such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methyl-2-pyrrolidone, dimethylsulfoxide or sulfolane, a halogenated hydrocarbon such as methylene chloride or chloroform, a nitrile such as acetonitrile or propionitrile, an ester such as ethyl acetate or ethyl propionate, an aliphatic hydrocarbon such as pentane, hexane, cyclohexane or heptane, a pyridine such as pyridine or picoline or water, or a solvent mixture thereof.

The base may, for example, be an inorganic base e.g. an alkali metal hydroxide such as sodium hydroxide or potassium hydroxide, an alkaline earth metal hydroxide such as calcium hydroxide or magnesium hydroxide, an alkali metal carbonate such as sodium carbonate or potassium carbonate, or an alkali metal bicarbonate such as sodium hydrogen carbonate or potassium hydrogen carbonate, a metal hydride such as sodium hydride or potassium hydride, an alkoxide such as sodium methoxide, sodium ethoxide or potassium tert-butoxide, or an organic base such as triethylamine, N,N-dimethylaniline, pyridine, 4-N,N-dimethylaminopyridine or 1,8-diazabicyclo[5.4.0]-7-undecene.

The reaction temperature is an optional temperature within a range of from -30°C to the reflux temperature in the reaction system, preferably from 0 to 150°C. The reaction time varies depending upon the particular compound, but can be set within a range of from 10

minutes to 20 hours.

Process 4



[III] [I]

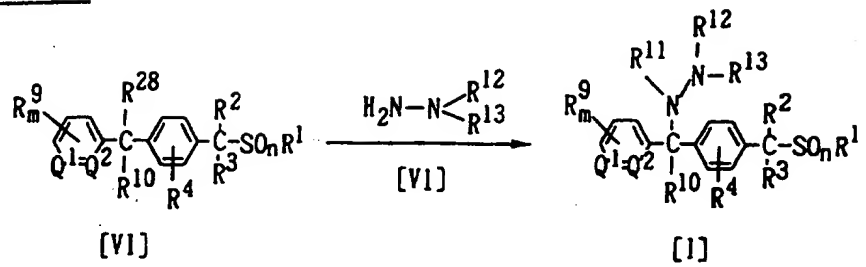
In the above formulas, R^1, R^2, R^3, R^4, R^9, R^{12}, R^{13}, m, Q^1 and Q^2 are as defined above; R^{29} is a hydroxyl group; and Z is a group of the formula $=SSR^1$.

In Process 4, a compound of the formula (I) of the present invention wherein A is (A2), can be obtained by reacting 1 mol of a benzyl alcohol of the formula (III) with from 1.0 to 3.0 mols of diaminochlorophosphine in from 0.1 to 5 ℓ of a solvent in the presence of from 1.0 to 3.0 mols of a base to obtain a phosphite, and then reacting it with from 1.0 to 5.0 mols of a disulfide of the formula (V2) in the presence of from 0 to 5 ℓ of a solvent.

The solvent and the base which can be used in this process may be the same as used in Process 3.

The reaction temperature is an optional temperature within a range of from -40°C to the reflux temperature in the reaction system, preferably from -30 to 50°C. The reaction time varies depending upon the particular compound, but can be set within a range of from 10 minutes to 20 hours.

Process 5



[VI] [I]

In the above formulas, R^1, R^2, R^3, R^4, R^9, R^{10}, R^{12}, R^{13}, m, n, Q^1 and Q^2 are as defined above; R^{11} is a hydrogen atom; and R^{28} is a halogen atom.

In Process 5, a compound of the formula (I) of the present invention wherein A is (A1), can be obtained by reacting 1 mol of a compound of the formula (VI) with from 1.0 to 10.0 mols of a hydrazine of the formula (V1) or its hydrate in the presence of from 0 to 5 ℓ of a solvent, if necessary in the presence of from 1.0 to 3.0 mols of a base.

The solvent which can be used, may, for example, be an ether such as diethyl ether, tetrahydrofuran or dioxane, an aromatic hydrocarbon such as benzene, toluene, xylene or chlorobenzene, an aprotic polar solvent such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methyl-2-pyrrolidone, dimethylsulfoxide or sulfolane, a halogenated hydrocarbon such as methylene chloride or chloroform, a nitrile such as acetonitrile or propionitrile, an ester such as ethyl acetate or ethyl propionate, an aliphatic hydrocarbon such as pentane, hexane, cyclohexane or heptane, a

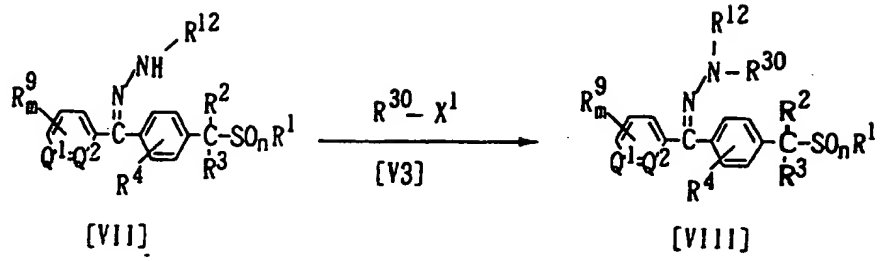
pyridine such as pyridine or picoline, or water, or a solvent mixture thereof.

The base may, for example, be an organic base e.g. an alkali metal hydroxide such as sodium hydroxide or potassium hydroxide, an alkaline earth metal hydroxide such as calcium hydroxide or magnesium hydroxide, an alkali metal carbonate such as sodium carbonate or potassium carbonate, an alkali metal bicarbonate such as sodium hydrogen carbonate or potassium hydrogen carbonate, a metal hydride such a sodium hydride or potassium hydride, an alkoxide such as sodium methoxide, sodium ethoxide or potassium tert-butoxide, or an organic base such as triethylamine, N,N-dimethylaniline, pyridine, 4-N,N-dimethylaminopyridine or 1,8-diazabicyclo[5.4.0]-7-undecene.

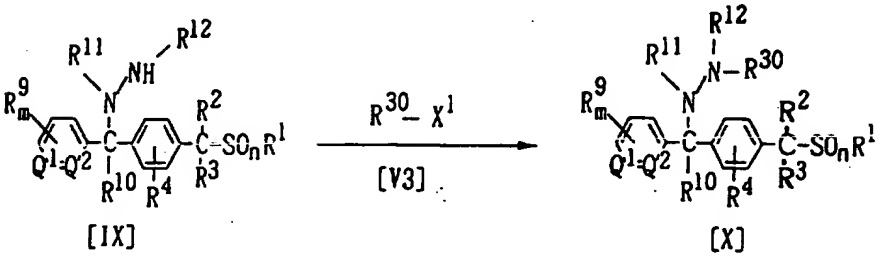
The reaction temperature is an optional temperature within a range of from -30°C to the reflux temperature in the reaction system, preferably from 0 to 150°C. The reaction time varies depending upon the particular compound, but can be set within a range of from 10 minutes to 20 hours.

The compound of the formula (I) of the present invention can also be produced by using the compound of the of the formula (I) of the present invention itself as the starting material. Such processes will be shown as Processes 6 to 11. However, such processes are not limited to these illustrated ones.

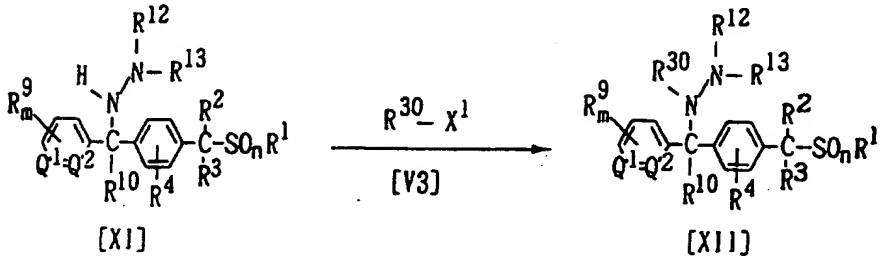
Process 6



[VII] [V3] [VIII]



[IX] [V3] [X]



[XI] [V3] [XII]

In the above formulas, R^{30} is a cyano group, a C_{1-6} alkyl group, a C_{1-4} haloalkyl group, a C_{2-10} alkoxyalkyl group, a C_{3-8} alkoxyalkoxyalkyl group, a C_{2-6} alkylthioalkyl group, a C_{2-6} alkenyl group, a C_{2-4} alkenyl group, a benzyl group (which may be substituted by a halogen atom, a methyl group or a trifluoromethyl group), a group of the formula $-COR^{14}$, a group of the formula $-COOR^{15}$, a group of the formula $-CON(R^{16})R^{17}$, a group of the formula $-SN-(R^{18})R^{19}$, a group of the formula $-SO_2R^{20}$,

a group of the formula $-C(R^{21})=CHR^{22}$ or a group of the formula $-C(R^{23})=NR^{25}$; when R^{30} is a group of the formula $-C(R^{21})=CHR^{22}$, X^1 is a halogen atom, a hydroxyl group, a C_{1-4} alkoxy group, a C_{1-4} alkylcarbonyloxy group, a mercapto group, a C_{1-4} alkylthio group, a C_{1-4} alkylsulfonyloxy group or a benzenesulfonyl group (which may be substituted by a methyl group), and in other cases, X^1 is a halogen atom, a C_{1-4} alkylsulfonyloxy group or a benzenesulfonyloxy group (which may be substituted by a methyl group), or R^{30}-X^1 may form $R^{17}NCO$ or $ClSO_2NCO$; and R^1, R^2, R^3, R^4, R^9, R^{10}, R^{11}, R^{12}, R^{13}, R^{14}, R^{15}, R^{16}, R^{17}, R^{18}, R^{19}, R^{20}, R^{21}, R^{22}, R^{23}, R^{25}, Q^1, Q^2, m and n are as defined above.

Namely, a new compound (VIII), (X) or (XII) of the present invention can be obtained by reacting 1 mol of a compound (VII), (IX) or (XI) of the present invention with from 1.0 to 10.0 mols of a compound of the formula (V3) in the presence of from 0 to 5 ℓ of a solvent, if necessary, in the presence of from 0.1 to 3.0 mols of an acid or a base.

The solvent and the acid catalyst which can be used, may be the same as used in Process 1.

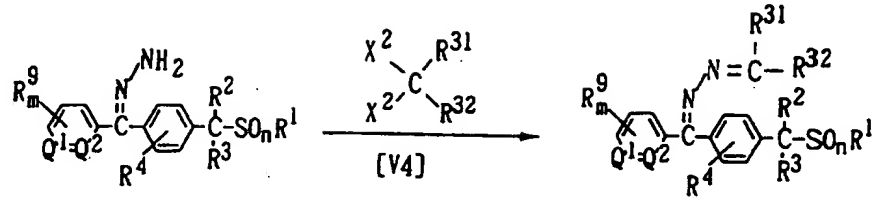
The base may, for example, be an inorganic base e.g. an alkali metal hydroxide such as sodium hydroxide or potassium hydroxide, an alkaline earth metal hydroxide such as calcium hydroxide or magnesium hydroxide, an alkali metal carbonate such as sodium carbonate or

potassium carbonate, or an alkali metal bicarbonate such as sodium hydrogen carbonate or a potassium hydrogen carbonate, a metal hydride such as sodium hydride or potassium hydride, an alkoxide such as sodium methoxide, sodium ethoxide or potassium tert-butoxide, or an organic base such as triethylamine, N,N-dimethylaniline, pyridine, 4-N,N-dimethylaminopyridine or 1,8-diazabicyclo[5.4.0]-7-undecene.

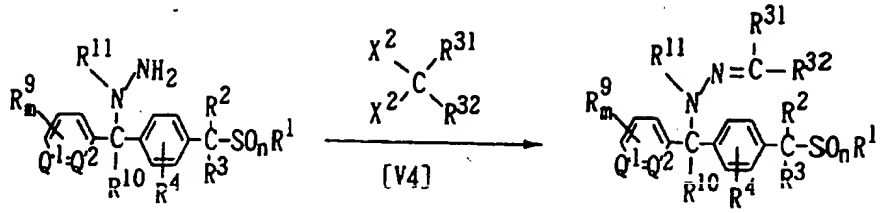
The reaction temperature is an optional temperature within a range of from -30°C to the reflux temperature in the reaction system, preferably from 0 to 150°C. The reaction time varies depending upon the particular compound, but can be set within a range of from 10 minutes to 20 hours.

When chlorosulfonyl isocyanate is reacted with the above compound (VII), (IX) or (XI), the resulting reaction product may be hydrolyzed after isolation or without isolation, to obtain a compound of the present invention wherein R^{30} is $CONH^2$.

Process 7



[XIII] [V4] [XV]



[XIV] [V4] [XVI]

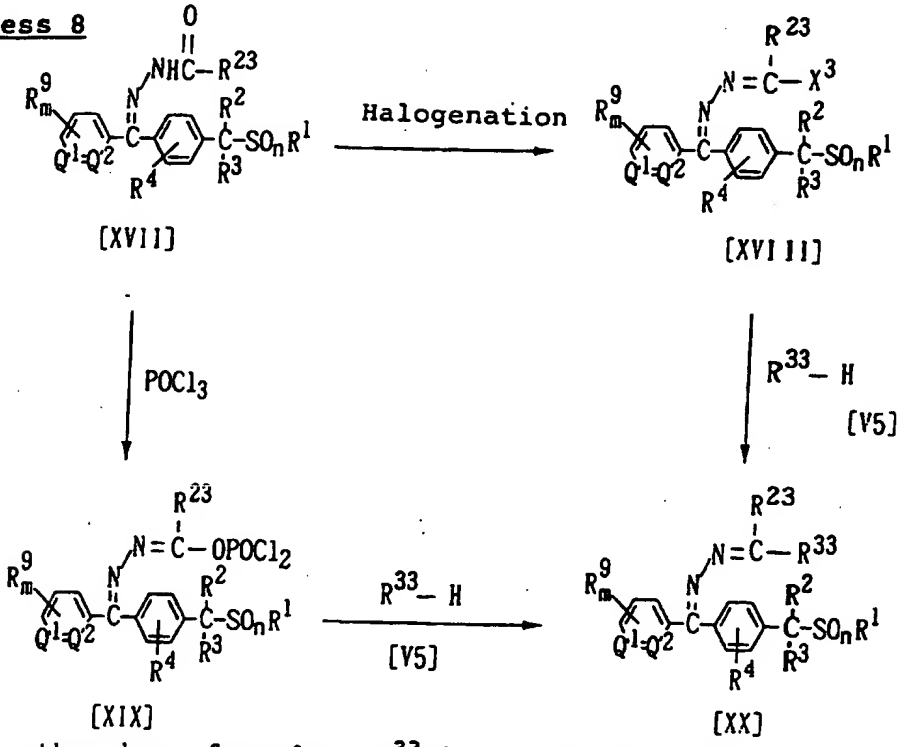
In the above formulas, each of R^{31} and R^{32} which are independent of each other, is a hydrogen atom, a C_{1-6} alkyl group or a group of the formula $-N(R^{25})R^{26}$; R^1, R^2, R^3, R^4, R^9, R^{10}, R^{11}, m, n, Q^1 and Q^2 are as defined above; each of R^{25} and R^{26} which are independent of each other, is a C_{1-4} alkyl group; X^2 is a C_{1-4} alkoxy group; or two X^2 may form a carbonyl group together with the carbon atom.

Namely, a new compound (XV) or (XVI) of the present invention can be obtained by reacting 1 mol of a compound of the formula (XIII) or (XIV) of the present invention with from 1.0 to 10.0 mols of a compound of the formula (V4) in the presence of from 0 to 5 ℓ of a solvent, if necessary in the presence of from 0.01 to 1.0 mol of an acid catalyst and a solvent.

This reaction can be conducted under the same

conditions as in Process 1.

Process 8



[XVII] [XVIII] [XIX] [XX]

In the above formulas, R^{33} is an azolyl group or a group of the formula $-N(R^{25})R^{26}$; R^1, R^2, R^3, R^4, R^9, R^{25}, R^{26}, m, n, Q^1 and Q^2 are as defined above; R^{23} is a hydrogen atom or a C_{1-6} alkyl group; and X^3 is a chlorine atom or a bromine atom.

Namely, a new compound (XVIII) of the present invention can be obtained by reacting 1 mol of a compound of the formula (XVII) of the present invention with from 1.0 to 10.0 mols of a halogenating agent in the presence of from 0 to 5 ℓ of a solvent. Then, from 1.0 to 5.0 mols of a compound of the formula (V5) is reacted in the presence of from 0 to 5 ℓ of a solvent, if necessary in the presence of from 1.0 to 3.0 mols of a base, to obtain

a new compound (XX) of the present invention. Further, the compound of the formula (XX) of the present invention can be prepared also by using a compound of the formula (XIX) instead of the compound of the formula (XVIII).

The halogenating agent may, for example, be phosphorus pentachloride, thionyl chloride, a mixture of triphenylphosphine/carbon chloride, or a mixture of triphenylphosphine/bromine.

The solvent which can be used, may, for example, be an ether such as diethyl ether, tetrahydrofuran or dioxane, an aromatic hydrocarbon such as benzene, toluene, xylene or chlorobenzene, a halogenated hydrocarbon such as methylene chloride or chloroform, a nitrile such as acetonitrile or propionitrile, an aliphatic hydrocarbon such as pentane, hexane, cyclohexane or heptane, or a solvent mixture thereof. Further, the halogenating agent may also serve as a solvent.

The reaction temperature is an optional temperature within a range of from 0°C to the reflux temperature in the system, preferably from 10 to 180°C. The reaction time varies depending upon the particular compound, but can be set within a range of from 10 minutes to 20 hours.

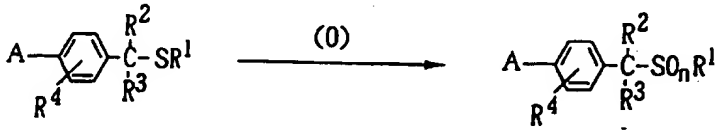
The compound of the formula (XIX) can be obtained by reacting a compound of the formula (XVII) with phosphorus oxychloride. A specific example of such a reaction is disclosed, for example, in Chemical Abstract, vol. 113,

97192b.

The compound of the formula (XX) of the present invention can usually be obtained by reacting a compound of the formula (XVIII) or (XIX) with a compound of the formula (V5) in the presence of a solvent, if necessary in the presence of a base and a catalyst.

The solvent and the base which can be used, may be the same as in Process 6. As the catalyst, a sulfinate such as sodium methanesulfinate or sodium p-toluenesulfinate, or its hydride, may be employed. The reaction temperature is an optional temperature within a range of from 0°C to the reflux temperature in the reaction system, preferably from 10 to 100°C. The reaction time varies depending upon the particular compound, but can be set within a range of from 10 minutes to 20 hours.

Process 9



[XXI] [XXII]

In the above formulas, n is 1 or 2; and A, R^1, R^2, R^3, R^4 and 1 are as defined above.

Namely, a new compound of the formula (XXII) of the present invention can be obtained by reacting 1 mol of a compound of the formula (XXI) of the present invention

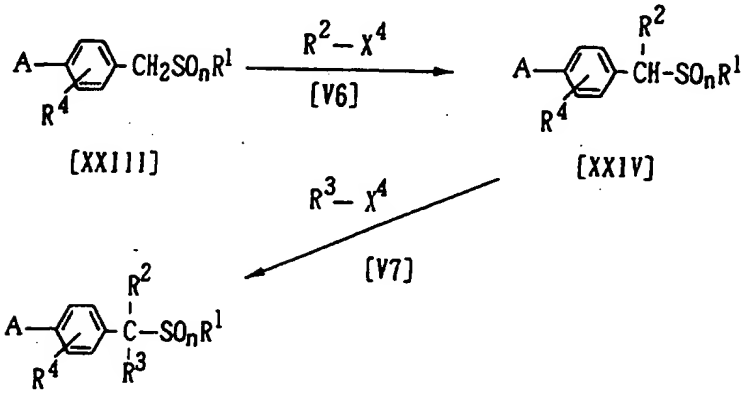
with from 1.0 to 10.0 mols of an oxidizing agent in the presence of from 0 to 5 ℓ of a solvent, if necessary in the presence of from 0.01 to 1.0 mol of a catalyst.

The oxidizing agent may, for example, be hydrogen peroxide, n-chloroperbenzoic acid, sodium periodate, OXONE (tradename for an agent containing potassium hydrogen peroxosulfate, manufactured by E.I. DuPont), N-chlorosuccinimide, N-bromosuccinimide, tert-butyl hypochlorite or sodium hypochlorite. The catalyst may, for example, be a sodium tungstate.

The solvent which can be used here, may, for example, be an ether, an aromatic hydrocarbon, an aprotic polar solvent, an alcohol, a halogenated hydrocarbon or an aliphatic hydrocarbon, as used in Process 1, acetic acid, water or a ketone such as acetone, methyl ethyl ketone or cyclohexanone, or a solvent mixture thereof.

The reaction temperature is an optional temperature within a range of from -20°C to the reflux temperature in the reaction system, preferably from 10 to 100°C. The reaction time varies depending upon the particular compound, but can be set within a range of from 10 minutes to 20 hours.

Process 10

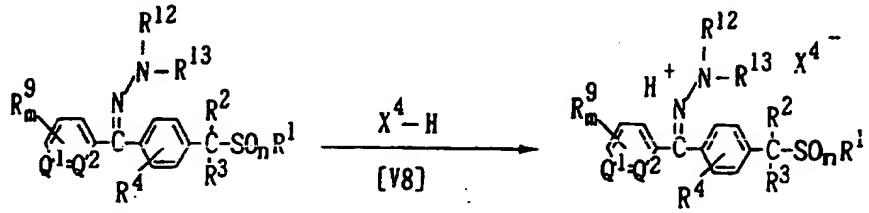


In the above formulas, each of R^2 and R^3 which are independent of each other, is a C_{1-4} alkyl group or a C_{1-3} haloalkyl group; A, R^2, R^4 and n are as defined above; and R^4 is a halogen atom, a C_{1-4} alkylsulfonyloxy group or a benzenesulfonyloxy group (which may be substituted by a methyl group).

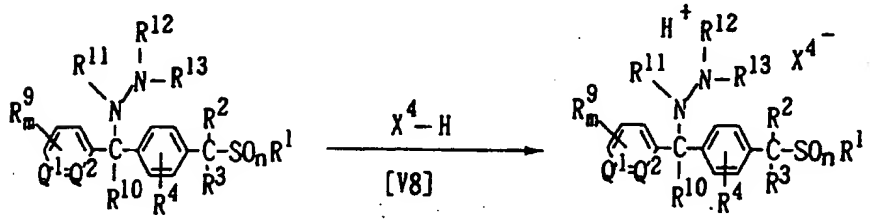
Namely, a compound of the formula (XXIV) can be produced by reacting 1 mol of a compound of the formula (XXIII) with from 1.0 to 5.0 mols of an alkylating agent of the formula (V6) in the presence of from 0 to 5 ℓ of a solvent and from 1.0 to 3.0 mols of a base. Then, this compound (XIV) may be reacted with from 1.0 to 5.0 mols of an alkylating agent of the formula (V7) after isolation or without isolation in the presence of from 0 to 5 ℓ of a solvent and from 1.0 to 3.0 mols of a base, to obtain a compound of the formula (XV).

The base and the solvent which can be used in these reactions, may, for example, be the same as used in Process 6. The reaction temperature is an optional temperature within a range of from -30°C to the reflux temperature in the reaction system, preferably from 0 to 100°C.

Process 11



[XXVI] [V8] [XXVII]



[XXVIII] [V8] [XXIX]

In the above formulas, X^4 is a halogen atom, a C_{1-4} alkylsulfonyloxy group or a benzenesulfonyloxy group (which may be substituted by a methyl group); and R^1, R^2, R^3, R^4, R^9, R^{10}, R^{11}, R^{12}, R^{13}, Q^1, Q^2, m and n are as defined above.

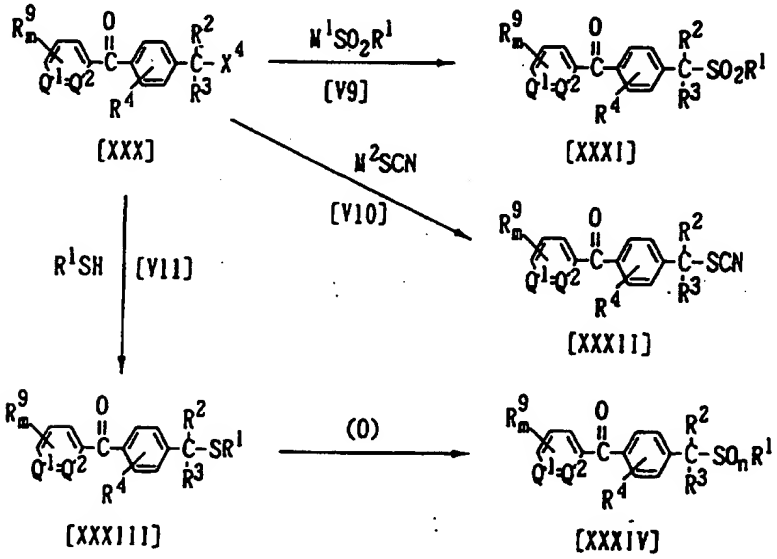
Namely, a salt of the compound (XVII) or (XIX) of the present invention can be obtained by reacting 1 mol of a compound of the formula (XXVI) or (XXVIII) of the present invention with from 1.0 to 3.0 mols of an acid of the

formula (V8) in the presence of from 0.1 to 5 ℓ of a solvent.

The solvent which can be used in such a reaction, may, for example, be the same as used in Process 1. The reaction temperature is an optional temperature within a range of from -30°C to the reflux temperature in the reaction system, preferably from 0 to 100°C.

The novel intermediates of the above formulas (II) and (III) can be produced, for example in accordance with the following Processes 12 to 20.

Process 12



In the above formulas, R^1 is a C_{1-6} alkyl group, a C_{1-4} cyanoalkyl group, a C_{1-4} hydroxylalkyl group, a C_{3-6} cycloalkyl group, a C_{1-6} haloalkyl group, a phenyl group (which may be substituted by a halogen atom or a C_{1-4} alkyl group), a benzyl group (which may be substituted by a halogen atom) or a thiazolyl group; X^4 is a halogen

atom, a C_{1-4} alkylsulfonyloxy group or a benzenesulfonyloxy group (which may be substituted by a methyl group); M^1 is an alkali metal; M^2 is an alkali metal or an ammonium ion; R^2, R^3, R^4, m, Q^1 and Q^2 are as defined above; and n is 1 or 2.

Namely, a benzyl sulfone derivative of the formula (XXXI) can be obtained by reacting 1 mol of a benzyl halide of the formula (XXX) with from 1.0 to 3.0 mols of an alkali metal salt of sulfinic acid of the formula (V9) in the presence of from 0 to 10 ℓ of a solvent.

The solvent which can be used here, may, for example, be an ether, an aromatic hydrocarbon, an aprotic polar solvent, an alcohol, a halogenated hydrocarbon, an aliphatic hydrocarbon or water, or a solvent mixture thereof. The reaction temperature is an optional temperature within a range of from 0°C to the reflux temperature in the reaction system, preferably from 10 to 100°C.

The salt of sulfinic acid to be used here, may be available as a reagent or can be prepared by a conventional method (e.g. a method disclosed in J. Chem. Soc., vol. 636 (1945), or J. A. Chem. Soc., vol 96, No. 7, p. 2275 (1974)).

Further, under similar reaction conditions, a compound of the formula (XXXII) can be obtained from a compound of the formula (XXX) and a thiocyanate of the formula (V10).

Furthermore, a sulfide of the formula (XXXIII) can be obtained by reacting 1 mol of the compound of the formula (XXX) with from 1.0 to 3.0 mols of a mercaptan of the formula (Vll) in the presence of from 0 to 5 ℓ of a solvent in the presence of from 1.0 to 0.3 mol of a base.

The solvent and the base may, for example, be the same as used in Process 6. The reaction temperature is an optional temperature within a range of from -10°C to the reflux temperature in the reaction system, preferably from 0 to 100°C.

A compound of the formula (XXXIV) of the present invention can be obtained by reacting 1 mol of the compound of the formula (XXXIII) thus obtained with from 1.0 to 10.0 mols of an oxidizing agent in the presence of from 0 to 5 ℓ of a solvent, if necessary in the presence of from 0.01 to 1.0 mol of a catalyst.

The oxidizing agent may, for example, be hydrogen peroxide, m-chloroperbenzoic acid, sodium periodate, OXONE (tradename for an agent containing potassium hydrogen peroxosulfate, manufactured by E.I. DuPont), N-chlorosuccinimide, N-bromosuccinimide, tert-butyl hypochlorite or sodium hypochlorite.

The catalyst may, for example, be sodium tungstate.

The solvent which can be used here, may, for example, be an ether, an aromatic hydrocarbon, an aprotic polar solvent, an alcohol, a halogenated hydrocarbon, an aliphatic hydrocarbon, as used in Process 1, acetic acid,

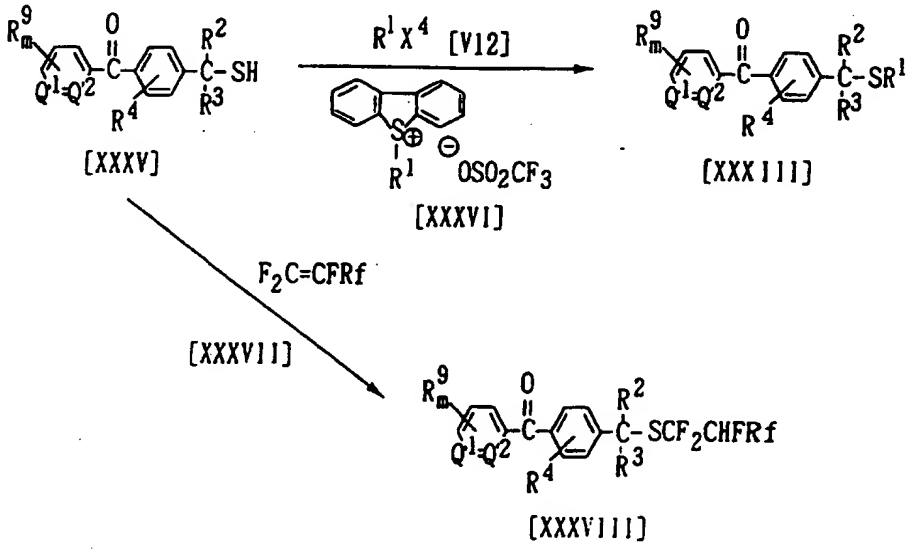
water or a ketone such as acetone, methyl ethyl ketone or cyclohexanone, or a solvent mixture thereof.

The reaction temperature is an optional temperature within a range of from -20°C to the reflux temperature in the reaction system, preferably from 10 to 100°C.

The benzyl halide of the formula (XXX) to be used as the starting material, is commonly known or can be prepared by a conventional method (e.g. a method disclosed in Org. Synth., vol. 4, p. 921 (1963)) by halogenating the methyl group of the corresponding arylcarbonyltoluene with a halogenating agent (such as chlorine, bromine, N-chlorosuccinimide, N-bromosuccinimide, sulfuryl chloride or sulfuryl bromide).

The arylcarbonyltoluene can be obtained usually by reacting toluene with an aryl carboxylic acid halide in the presence of a Lewis acid such as aluminum chloride.

Process 13



In the above formulas, R^1 is a C_{1-6} alkyl group, a C_{3-6} cycloalkyl group, a C_{1-6} haloalkyl group, a C_{1-4} cyanoalkyl group or a C_{1-4} hydroxyalkyl group; R_f is a fluorine atom or a perfluoroalkyl group; X^4 is a halogen atom, a C_{1-4} alkylsulfonyloxy group or a benzenesulfonyloxy group (which may be substituted by a methyl group); R^2, R^3, R^4, m, Q^1 and Q^2 are as defined above; and n is 1 or 2.

The sulfide of the formula (XXXIII) or (XXXVIII) can be obtained by reacting 1 mol of benzyl mercaptan of the formula (XXXV) with from 1.0 to 3.0 mols of an alkylating agent of the formula (Vl2) and a dibenzothiopheniumtrifluoromethane sulfonate of the formula (XXXVI), or a perfluoroalkene of the formula (XXXVII), in the presence of from 0 to 10 ℓ of a solvent, if necessary in the presence of from 1.0 to 3.0 mols of a base.

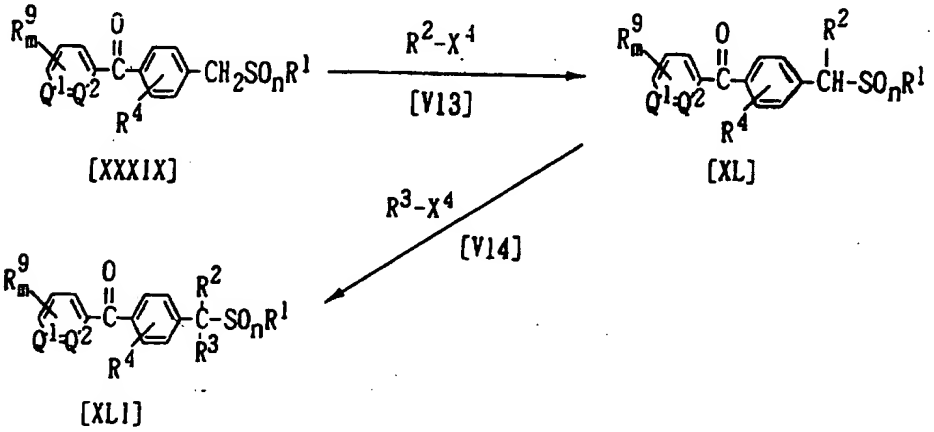
The base and the solvent which can be used here, may, for example, be the same as used in Process 6. The reaction temperature is an optional temperature within a range of from 0°C to the reflux temperature in the reaction system, preferably from 10 to 150°C.

By oxidizing the sulfide of the formula (XIII) or (XVIII) thus obtained by the same oxidizing method as used in Process 12, it is possible to obtain the corresponding sulfoxide or sulfone derivative.

The benzyl mercaptan of the formula (XXXV) to be used

as the starting material, is already known or can be prepared in accordance with a conventional method (e.g. a method disclosed in Org. Synth., vol. 3, p. 363 (1955)) or a similar method. Namely, it can be obtained by reacting a benzyl halide of the formula (XXX) as the starting material in Process 12, with sodium sulfide, or reacting it in the presence of thiourea and a base, followed by hydrolysis.

Process 14



[XXXIX] [XL] [XLI]

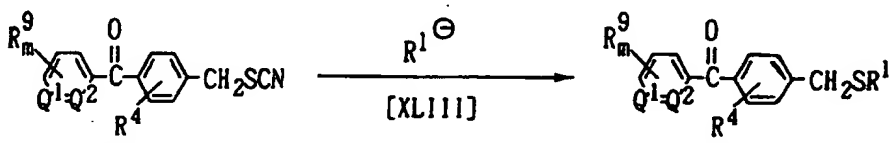
In the above formulas, R^1, R^4, R^9, m, n, Q^1 and Q^2 are as defined above; each of R^2 and R^3 is an alkyl group or a haloalkyl group; and X^4 is a halogen atom, a C_{1-4} alkylsulfonyloxy group or a benzene sulfonyloxy group (which may be substituted by a methyl group).

Namely, the compound of the formula (XL) or (XVI) can be prepared by reacting 1 mol of a compound of the formula (XXXIX) with from 1.0 to 5.0 mols of an alkylating agent of the formula (V13) or (V14) in the presence of from 0 to 5 ℓ of a solvent and from 1.0 to

3.0 mols of a base. When X^4 is present in a side chain of R^1, R^1 and R^2 will form a from 3- to 8-membered ring having one or more hetero atoms, together with the sulfur and carbon atoms to which they are respectively bonded.

The base and the solvent which can be used here, may, for example, be the same as used in Process 6. The reaction temperature is an optional temperature within a range of from 0°C to the reflux temperature in the reaction system, preferably from 10 to 150°C.

Process 15



[XLII] [XLIII] [XLIV]

In the above formulas, R^4, R^9, m, n, Q^1 and Q^2 are as defined above; and R^1 is an alkyl group or a haloalkyl group.

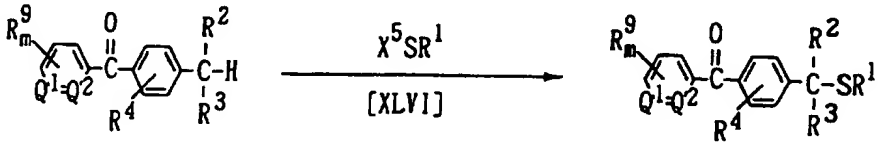
Namely, a compound of the formula (XLIV) can be produced by reacting 1 mol of the compound of the formula (XLII) of the present invention with from 1.0 to 5.0 mols of a carbanion of the formula (XLIII) in the presence of from 0 to 10 ℓ of a solvent. The method for generating the carbanion of the formula (XLIII), may, for example, be 1) a method of contacting from 1.0 to 15.0 mols of a trihalomethane with from 1.0 to 15.0 mols of a base, if necessary in the presence of from 0.01 to 1.0 mol of a

phase transfer catalyst such as a tetraalkyl ammonium salt, a benzyltrialkylammonium salt, a tetraalkylphosphonium salt or a crown ether, 2) a method of contacting from 1.0 to 15.0 mols of a (trialkylsilyl)alkyl halide with from 1.0 to 15.0 mols of a hydrofluoride such as potassium fluoride or tetrabutylammonium fluoride, or 3) a method of contacting from 1.0 to 5.0 mols of an alkyl halide or a haloalkyl halide with from 1.0 to 5.0 mols of a metal such as lithium, sodium, copper or zinc, or with an organometallic compound such as lithium diisopropylamide, phenyl lithium or butyl lithium.

The solvent which can be used, may, for example, be an ether, an aromatic hydrocarbon, an aprotic polar solvent, an alcohol, an aliphatic hydrocarbon or water, or a solvent mixture thereof.

The reaction temperature is an optional temperature within a range of from -70°C to the reflux temperature in the reaction system, preferably from -50 to 50°C.

Process 16



[XLV] [XLVII]

In the above formula, R^2, R^3, R^4, R^9, m, n, Q^1 and Q^2 are as defined above; X^5 is a halogen atom, a cyano group or a group of the formula SR^1; and R^1 is an alkyl group

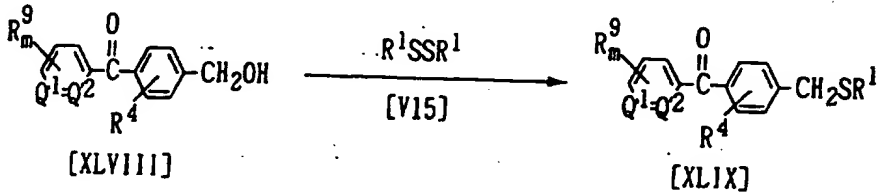
or a haloalkyl group.

Namely, a compound of the formula (XLVII) can be prepared by reacting 1 mol of a compound of the formula (XLV) of the present invention with from 1.0 to 5.0 mols of a compound of the formula (XLVI) in the presence of from 0 to 5 ℓ of a solvent and from 1.0 to 5.0 mols of a base.

The base and the solvent which can be used here, may, for example, be the same as used in Process 6.

The reaction temperature is an optional temperature within a range of from -70°C to the reflux temperature in the reaction system, preferably from -50 to 50°C.

Process 17



[XLVIII] [XLIX]

In the above formulas, R^1, R^4, R^9, m, n, Q^1 and Q^2 are as defined above.

Namely, a compound of the formula (XLIX) of the present invention can be obtained by reacting 1 mol of a benzyl alcohol of the formula (XLVIII) with from 1.0 to 3.0 mol of a dialkylaminochlorophosphine usually in the presence of a solvent and a base, i.e. in the presence of from 0.1 to 5 ℓ of a solvent and from 1.0 to 3.0 mols of a base, to obtain a phosphite, followed by reacting from

1.0 to 5.0 mols of a disulfide of the formula (V15) in the presence of from 0 to 5 ℓ of a solvent.

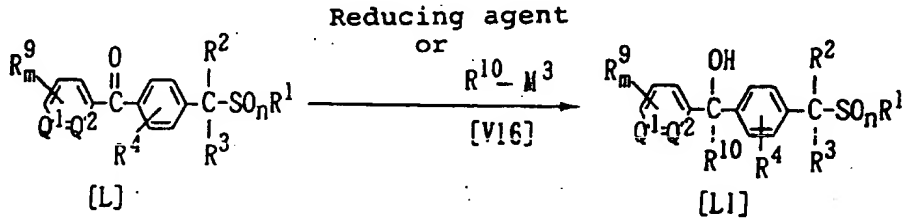
The solvent which can be used here, may, for example, be an ether such as diethyl ether, tetrahydrofuran or dioxane, an aromatic hydrocarbon such as benzene, toluene, xylene or chlorobenzene, an aprotic polar solvent such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methyl-2-pyrrolidone, dimethylsulfoxide or sulfolane, a halogenated hydrocarbon such as methylene chloride or chloroform, a nitrile such as acetonitrile or propionitrile, an ester such as ethyl acetate or ethyl propionate, an aliphatic hydrocarbon such as pentane, hexane, cyclohexane or heptane, a pyridine such as pyridine or picoline, or a solvent mixture thereof.

The base may, for example, be an inorganic base e.g. an alkali metal hydroxide such as sodium hydroxide or potassium hydroxide, an alkaline earth metal hydroxide such as calcium hydroxide or magnesium hydroxide, an alkali metal carbonate such as sodium carbonate or potassium carbonate, or an alkali metal bicarbonate such as sodium hydrogen carbonate or potassium hydrogen carbonate, a metal hydride such as sodium hydride or potassium hydride, or an organic base such as triethylamine, N,N-dimethylaniline, pyridine, 4-N,N-dimethylaminopyridine or 1,8-diazabicyclo[5.4.0]-7-undecene.

The reaction temperature is an optional temperature within a range of from -40°C to the reflux temperature in the reaction system, preferably from -30 to 50°C.

The reaction time varies depending upon the particular compound, but can be set within a range of from 10 minutes to 20 hours.

Process 18



In the above formulas, R^1, R^2, R^3, R^4, R^9, R^{10}, Q^1, Q^2, M^3, m and n are as defined above.

Namely, a compound of the formula (LI) of the present invention can be obtained by reacting a compound of the formula (L) of the present invention with from 1.0 to 50.0 mols of a reducing agent in the presence of from 0 to 5 ℓ of a solvent, if necessary, in the presence of from 0.01 to 1.0 mol of a catalyst, or by reacting it with from 1.0 to 5.0 mols of an alkyl metal compound of the formula (V16).

The reducing agent may, for example, be molecular hydrogen, sodium borohydride, lithium aluminum hydride or aluminum hydride or diisobutylaluminum hydride.

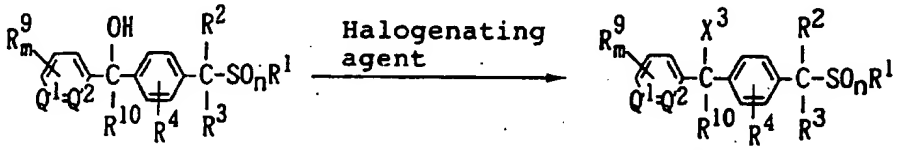
The catalyst may, for example, be platinum, nickel, cobalt or palladium.

The solvent which can be used here, may, for example,

be an ether, an aromatic hydrocarbon, an aprotic polar solvent, an alcohol, an aliphatic hydrocarbon, acetic acid or water, or a solvent mixture thereof.

The reaction temperature is an optional temperature within a range of from -20°C to the reflux temperature in the reaction system, preferably from 10 to 100°C. The reaction time varies depending upon the particular compound, but can be set within a range of from 10 minutes to 20 hours.

Process 19



[LI] [LII]

In the above formulas, R^1, R^2, R^3, R^4, R^9, R^{10}, Q^1, Q^2, m and n are as defined above; and X^3 is a chlorine atom or a bromine atom.

A new compound of the formula (LII) of the present invention can be obtained by reacting 1 mol of a compound of the formula (LI) of the present invention with from 1.0 to 10.0 mols of a halogenating agent in the presence of from 0 to 5 ℓ of a solvent.

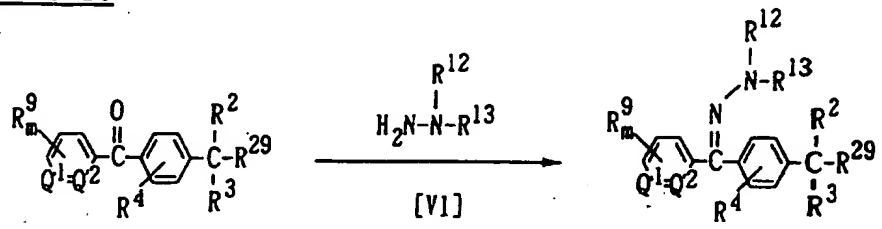
The halogenating agent may, for example, be hydrogen chloride, hydrogen bromide, phosphorus trichloride, phosphorus tribromide, thionyl chloride, a mixture of triphenylphosphine/carbon tetrachloride or a mixture of a

triphenylphosphine/bromine.

The solvent which can be used here, may, for example, be an ether such as diethyl ether, tetrahydrofuran or dioxane, an aromatic hydrocarbon such as benzene, toluene, xylene or chlorobenzene, a halogenated hydrocarbon such as methylene chloride or chloroform, a nitrile such as acetonitrile or propionitrile, an aliphatic hydrocarbon such as pentane, hexane, cyclohexane or heptane, or a solvent mixture thereof. Further, the halogenating agent may serve as a solvent.

The reaction temperature is an optional temperature within a range of from 0°C to the reflux temperature in the reaction system, preferably from 10 to 180°C. The reaction time varies depending upon the particular compound, but can be set within a rang of from 10 minutes to 20 hours.

Process 20



[LIII] [VI] [III]

In the above formulas, R^2, R^3, R^4, R^9, R^{12}, R^{13}, R^{29}, Q^1, Q^2 and m are as defined above.

Namely, a compound of the formula (III) of the present invention can be obtained by reacting 1 mol of a benzophenone of the formula (LIII) with from 1.0 to 10.0

mols of a hydrazine of the formula (VI) or its hydrate in the presence of from 0 to 5 ℓ of a solvent, if necessary in the presence of from 0.01 to 1.0 mol of an acid catalyst.

The solvent and the acid catalyst which can be used here, may, for example, be the same as used in Process 1.

The reaction temperature is an optional temperature within a range of from -10°C to the reflux temperature in the reaction system, preferably from 0 to 100°C.

BEST MODE FOR CARRYING OUT THE INVENTION

Now, the presence invention will be described in further detail with reference to Preparation Examples, Formulation Examples and Test Examples.

PREPARATION EXAMPLE 1

Preparation of 4-chloro-4'-trifluoromethylsulfonylmethylbenzophenone-N'-ethoxycarbonylhydrazone (Compound No. I-175)

4-chloro-4'-trifluoromethylsulfonylmethylbenzophenone (0.5 g) and ethyl-carbazate (0.4 g) were added to ethanol (40 ml) and acetic acid (5 ml), and the mixture was stirred for 19 hours under reflux. The reaction mixture was concentrated, and the residue was extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=4:1) to obtain the

desired product (0.6 g, melting point: 148-150°C, yield: 96%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

1.30	(3H, t)
4.23	(2H, q)
4.35, 4.53	(2H, s, s)
7.03-7.80	(9H, m)

PREPARATION EXAMPLE 2

Preparation of 4-chloro-4'-methylsulfonylmethyl-benzophenone-hydrazone (Compound No. I-2)

4-chloro-4'-methylsulfonylmethylbenzophenone (10.0 g) and hydrazine monohydrate (4.9 g) were added to ethanol (200 ml) and acetic acid (10 ml), and the mixture was stirred for 6 hours under reflux. The reaction mixture was concentrated, and the residue was extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=2:1) to obtain the desired product (10.0 g, melting point: 52 to 54°C, yield: 97%)

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

2.73	(3H, s, s)
4.20, 4.30	(2H, s, s)
5.50	(2H, br)
7.05-7.70	(8H, m)

PREPARATION EXAMPLE 3

Preparation of 4-chloro-4'-methylsulfonylmethylbenzophenone-N'-propionylhydrazone (Compound No. I-15)

4-chloro-4'-methylsulfonylmethylbenzophenone-hydrazone (1.3 g), propionyl chloride (0.4 g) and potassium carbonate (0.7 g) were added to ethyl acetate (150 ml) and water (100 ml), and the mixture was stirred for 2 hours at room temperature. The reaction mixture was subjected to liquid separation, and the ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=2:1) to obtain the desired product (1.3 g, melting point: 159 to 160°C, yield: 86%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

1.23	(3H, t)
2.85-3.00	(5H, m)
4.30	(2H, s)
7.00-8.00	(8H, m)
8.25	(1H, br)

PREPARATION EXAMPLE 4

Preparation of 4-chloro-4'-trifluoromethylsulfonyl-methylbenzophenone (Compound No. I-136)

4-chloro-4'-trifluoromethylsulfonylmethylbenzophenone (2.5 g), hydrazine monohydrate (4.3 g) and p-toluene

sulfonic acid monohydrate (0.2 g) were added to ethanol (30 ml), and the mixture was stirred for 3 hours under reflux. The reaction mixture was concentrated, and the residue was extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=2:1) to obtain the desired product (2.2 g, n_D^{20}=1.5871, yield: 85%)

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

4.43, 4.53	(3H, s, s)
5.47, 5.53	(2H, s, s)
7.07-7.60	(8H, m)

PREPARATION EXAMPLE 5

Preparation of 4-chloro-4'-trifluoromethylsulfonyl-methylbenzophenone (Compound No. I-149)

4-chloro-4'-trifluoromethylsulfonylmethylbenzophenone (0.9 g), propionyl chloride (0.22 g) and potassium carbonate (0.4 g) were added to a solvent comprising ethyl acetate (100 ml) and water (100 ml), and the mixture was stirred for 16 hours at room temperature. The reaction mixture was subjected to liquid separation, and the ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residual solid was washed with n-hexane to obtain the

desired product (0.75 g, melting point: 130 to 132°C, yield: 75%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

1.20	(3H, t)
2.60-3.00	(2H, m)
4.47	(2H, d)
7.03-7.63	(8H, m)
8.22	(1H, d)

PREPARATION EXAMPLE 6

Preparation of 4-chloro-4'-trifluoromethylsulfonyl-methylbenzophenone'-(1-chloropropylidene)hydrazone (Compound No. II-14)

4-chloro-4'-trifluoromethylsulfonylmethyl-benzophenone-propionylhydrazone (1.7 g), triphenylphosphine (1.5 g) and carbon tetrachloride (1.2 g) were added to acetonitrile (80 ml), and the mixture was stirred for 10 minuted under reflux. The reaction mixture was concentrated, and the residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=10:1) to obtain the desired product (1.7 g, melting point: 108-109°C, yield: 97%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

1.05, 1.10	(3H, t, t)
2.50, 2.55	(2H, q, q)
4.47	(2H, s)
7.00-7.85	(8H, m)

PREPARATION EXAMPLE 7

Preparation of 4-chloro-4'-trifluoromethylsulfonyl-methylbenzophenone'-[1-(1-H-1,2,4-triazole-1-yl)propylidene]hydrazone (Compound No. II-10)

4-chloro-4'-trifluoromethylsulfonyl-methylbenzophenone'-(1-chloropropylidene)hydrazone (1.0 g), 1-H-1,2,4-triazole (0.2 g), potassium carbonate (0.4 g) and sodium p-toluene sulfonate (0.3 g) were added to N,N-dimethylformamide (70 ml), and the mixture was stirred for 7 hours at a temperature of from 95 to 100°C. The reaction mixture was cooled to room temperature, and water was added thereto. The mixture was extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=6:1) to obtain the desired product (0.7 g, n_D^{20}=1.5978, yield: 66%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

1.28	(3H, t)
3.28	(2H, q)
4.52	(2H, s)
7.05-7.86	(8H, m)
7.95	(1H, s)
8.40, 8.52	(1H, s, s)

PREPARATION EXAMPLE 8

Preparation of 4-chloro-4'-trifluoromethylsulfonyl-methylbenzophenone'-[1-(N-

methylamino)ethylidene]hydrazone (Compound No. II-8)

4-chloro-4'-trifluoromethylsulfonylmethyl-benzophenone'-(1-chloroethylidene)hydrazone (0.7 g) and a 40% methylamine aqueous solution (0.3 g) were added to xylene (50 ml), and the mixture was stirred for one hour under reflux. The reaction mixture was concentrated, and the residue was extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=1:1) to obtain the desired product (0.6 g, melting point: 58 to 60°C, yield: 87%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

2.00, 2.20	(3H, s, s)
2.67, 2.94	(3H, d, d)
4.46	(2H, s)
6.30	(1H, br)
6.95-7.78	(8H, m)

PREPARATION EXAMPLE 9

Preparation of 4-chloro-4'-ethylsulfonyl-methylbenzophenone-N'-isopropylidenehydrazone (Compound No. II-35)

4-chloro-4'-ethylsulfonylmethylbenzophenone hydrazone (0.7 g) was added to acetone (30 ml), and the mixture was stirred for 30 minutes under reflux. The reaction mixture was concentrated to obtain the desired product

(0.7 g, n_D^{20}=1.6163, yield: 88%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

1.34	(3H, t)
2.00	(6H, s)
2.90	(2H, q)
4.20	(2H, s)
7.00-7.67	(8H, m)

PREPARATION EXAMPLE 10

Preparation of 4-chloro-4'-methylthiomethylbenzophenone-N'-[1-(N,N-dimethylamino)ethylidene]hydrazone (Compound No. II-22)

4-chloro-4'-methylthiomethylbenzophenone-hydrazone (2.0 g) and methylacetamide-dimethylacetal (1.4 g) were added to ethanol (100 ml), and the mixture was stirred for 6 hours under reflux. The reaction mixture was concentrated and extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residue was purified by silica gel chromatography (n-hexane:ethyl acetate=6:1) to obtain the desired product as a slightly yellow transparent viscous liquid (2.0 g, yield: 81%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

2.00	(3H, s)
2.35	(3H, s)
2.88	(6H, s)
3.66, 3.70	(2H, s, s)

7.03-7.75 (8H, m)

PREPARATION EXAMPLE 11

Preparation of 4-chloro-4'-methylsulfonylmethylbenzophenone-semicarbazone (Compound No. I-42)

4-chloro-4'-methylsulfonylmethylbenzophenone-hydrazone (1.3 g) and chlorosulfonyl isocyanate (0.63 g) were added to ethyl acetate (100 ml), and the mixture was stirred for one hour at room temperature. Then, water (100 ml) was added, and the mixture was further stirred for 16 hours at room temperature. The reaction mixture was subjected to liquid separation, and the ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residual solid was washed with a solvent mixture of ethyl acetate:n-hexane=4:1, to obtain the desired product (1.2 g, melting point: 189 to 191°C, yield: 80%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

2.87, 2.97 (3H, s, s)

4.34, 4.50 (5H, m)

7.10-7.70 (8H, m)

PREPARATION EXAMPLE 12

Preparation of 4-chloro-4'-methylthiomethylbenzophenone-N'-ethoxycarbonyl-N'-methylhydrazone (Compound No. I-47)

4-chloro-4'-methylthiomethylbenzophenone-N'-ethoxycarbonylhydrazone (4.3 g) was dissolved in N,N-

dimethylformamide (100 ml). To this solution, 60% sodium hydride (0.6 g) was added, and the mixture was stirred for 30 minutes at room temperature. Then, methyl iodide (2.5 g) was added thereto, followed by stirring for 16 hours at room temperature. Water was added to the reaction mixture, and the mixture was extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=5:1) to obtain the desired product (4.3 g, n_D^{20}:1.6042, yield: 89%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

1.17	(3H, t)
2.00	(3H, s)
2.79, 3.00	(3H, s, s)
3.63, 3.67	(2H, s, s)
4.04	(2H, q)
7.07-7.57	(8H, m)

PREPARATION EXAMPLE 13

Preparation of 4-chloro-4'-trifluoromethylsulfonyl-methylbenzophenone-4-butylsemicarbazone (Compound No. I-171)

4-chloro-4'-trifluoromethylsulfonylmethyl-benzophenonehydrazone (1.2 g), triethylamine (0.5 g) and butyl isocyanate (0.6 g) were added to tetrahydrofuran (30 ml), and the mixture was stirred for 16 hours at room

temperature. The reaction mixture was concentrated, and the residue was extracted with ethyl acetate. The ethyl acetate layer was washed with 2N hydrochloric acid and water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residue was purified by silica gel chromatography (n-hexane:ethyl acetate=4:1) to obtain the desired product (0.6 g, melting point: 169 to 181°C, yield: 40%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

	0.75-1.65	(7H, m)
	3.15-3.50	(2H, m)
	4.50	(2H, s)
	6.20	(1H, br)
	6.90-7.70	(9H, m)

PREPARATION EXAMPLE 14

Preparation of 4-chloro-4'-trifluoromethylsulfonylmethylbenzophenone-N'-methylsulfonyliminomethylhydrazone (Compound No. I-137)

4-chloro-4'-trifluoromethylsulfonylmethylbenzophenonehydrazone (1.2 g) triethylamine (1.6 g) and N-methylsulfonylformimide acid ethyl (1.2 g) were added to dioxane (30 ml), and the mixture was stirred for 5 hours under reflux. The reaction mixture was concentrated, and the residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=1:1) to obtain the desired product (0.8 g, melting point: 63 to 65°C, yield: 52%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

2.95, 3.05	(3H, s, s)
4.45, 4.60	(2H, s, s)
4.95	(1H, br)
7.10-7.80	(8H, m)
8.80	(1H, br)

PREPARATION EXAMPLE 15

Preparation of 4-chloro-4'-trifluoromethylsulfonylmethylbenzophenone-N'-methylsulfonylhydrazone (Compound No. I-182)

4-chloro-4'-trifluoromethylsulfonylmethylbenzophenonehydrazone (1.2 g) and triethylamine (0.4 g) were dissolved in ethyl acetate (30 ml). To this solution, methane sulfonyl chloride (0.4 g) was dropwise added at room temperature, and the mixture was stirred for one hour. The reaction mixture was washed with 2N hydrochloric acid and water and then and dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=4:1) to obtain the desired product (0.5 g, melting point: 64 to 65°C, yield: 36%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

3.50	(3H, s)
4.50	(2H, d)
7.10-7.70	(9H, m)

PREPARATION EXAMPLE 16

Preparation of 4-chloro-4'-methylsulfinylmethylbenzophenone-N'-propionylhydrazone (Compound No. I-14)

4-chloro-4'-methylthiomethylbenzophenone-N'-propionylhydrazone (0.8 g) and sodium periodide (0.5 g) were added to methanol (50 ml) and water (7 ml), and the mixture was stirred for 16 hours at room temperature. The reaction mixture was concentrated, and the residue was extracted with ethyl acetate. The ethyl acetate layer was washed with water and dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=10:1) to obtain the desired product (0.7 g, melting point: 153 to 156°C, yield: 84%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

1.21	(3H, t)
2.43, 2.56	(3H, s, s)
2.85	(2H, q)
3.93, 4.00	(2H, s, s)
6.96-7.70	(8H, m)
8.23	(1H, br)

PREPARATION EXAMPLE 17

Preparation of 4-chloro-4'-(2-methylsulfonyl-2-propyl)benzophenone-N'-hexanolyl-N'-methylhydrazone (Compound No. I-128)

4-chloro-4'-methylsulfonylmethylbenzophenone-N'-hexanolylhydrazone (1.4 g), methyl iodide (5.6 g) and 60% sodium hydride (0.15 g) were added to N,N-dimethylformamide (80 ml), and the mixture was stirred for 16 hours at room temperature. To the reaction mixture, water was added, and the mixture was extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residue was purified by silica gel chromatography (n-hexane:ethyl acetate=2:1) to obtain the desired product (0.8 g, melting point: 94 to 96°C, yield: 53%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

0.90	(3H, m)
1.10-1.93	(6H, m)
1.87	(6H, s)
2.16-2.67	(2H, m)
2.57, 2.76	(3H, s)
3.09	(3H, s)
7.05-7.73	(8H, m)

PREPARATION EXAMPLE 18

Preparation of 4-chloro-4'-trifluoromethylsulfonylmethylbenzhydrylhydrazine (Compound No. V-8)

Hydrazine monohydrate (50 ml) and 4-chloro-4'-trifluoromethylsulfonylmethylbenzhydryl chloride were

added to toluene (80 ml), and the mixture was gradually heated with stirring. The mixture was stirred at 80°C for 2 hours, then left to cool and poured into water. 250 ml of ethyl acetate was added thereto for extraction, and the extract was washed with water and dried over anhydrous magnesium sulfate. Then, the solvent was distilled off under reduced pressure to obtain the product as a slightly yellow viscous substance (2.9 g, n_D^{20}: 1.5671, yield: 82.4%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

3.83	(2H, br)
4.38	(2H, s)
4.83	(1H, s)
6.96-7.50	(8H, m)

PREPARATION EXAMPLE 19

Preparation of N-(4-chloro-4'-trifluoromethylsulfonylmethylbenzhydryl)-N'-methoxycarbonylhydrazine hydrochloride (Compound No. V-13)

N-(4-chloro-4'-trifluoromethylsulfonylmethylbenzhydryl)-N'-methoxycarbonylhydrazine (1.6 g) was added to methanol (80 ml), and hydrochloric acid (3 ml) was added thereto with stirring at room temperature. The mixture was gradually heated to the reflux temperature and stirred for 2 hours under reflux, and then it was left to cool. The solvent was distilled off under reduced pressure to obtain the desired product as a

slightly yellow powder (1.6 g, melting point: 52 to 54°C, yield: 91.5%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

3.6	(3H, s)
4.73	(2H, s)
5.23	(1H, s)
7.27-7.50	(8H, m)

PREPARATION EXAMPLE 20

Preparation of 4-chloro-4'-ethylthiomethylbenzophenone-N'-ethoxycarbonylhydrazone (Compound No. I-114)

Ethane thiol (1.2 g) and sodium hydroxide (1 g) were suspended in N,N-dimethylformamide (50 ml), and then 4-chloro-4'-chloromethylbenzophenone-N'-ethoxycarbonylhydrazone (3.5 g) was added thereto. The mixture was stirred for 16 hours at room temperature. Water was added to the reaction mixture, and the mixture was extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=4:1) to obtain the desired product (2.0 g, n_D^{20}:1.6198, yield: 53%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

1.34	(6H, t)
2.53	(2H, q)
3.70, 3.80	(2H, q)

7.10-7.77 (8H, m)

PREPARATION EXAMPLE 21

Preparation of 4-chloro-4'-difluoromethylthiomethylbenzophenone-N'-methoxycarbonylhydrazone (Compound No. I-187)

4-chloro-4'-mercaptomethylbenzophenone-N'-methoxycarbonylhydrazone (1.5 g) and potassium hydroxide (1.5 g) were added to a solvent comprising dioxane (30 ml) and water (30 ml). Difluoromethyl chloride was blown into this solution at 40°C until the starting material 4-chloro-4'-mercaptomethylbenzophenone-N'-methoxycarbonylhydrazone disappeared. The reaction mixture was cooled to room temperature and subjected to filtration. The organic layer as the filtrate was dried over anhydrous magnesium sulfate and concentrated. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=4:1) to obtain the desired product (0.3 g, n_D^{20}:1.6213, yield: 18%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

3.78	(3H, s)
3.98, 4.10	(2H, s, s)
6.7, 6.8	(1H, t)
7.07-7.67	(8H, m)
7.77	(1H, s)

PREPARATION EXAMPLE 22

Preparation of 4-chloro-4'-methylthiomethylbenzophenone-N'-methoxycarbonylhydrazone (Compound No. I-67)

4-chloro-4'-hydroxymethylbenzophenone-N'-methoxycarbonylhydrazone (1.2 g) and triethylamine (0.5 g) were added to tetrahydrofuran (30 ml). Chlorobis(diethylaminophosphine) (1.1 g) was dropwise added to this solution at -20°C. The mixture was stirred for 2 hours at room temperature, and then the solvent was distilled off under reduced pressure. Ice water and ethyl acetate were added thereto for liquid separation. The organic layer was dried over anhydrous magnesium sulfate and concentrated to obtain a phosphite. This phosphite was added to tetrahydrofuran (30 ml). Dimethyl sulfide (0.9 g) was dropwise added to this solution at 0°C, and the mixture was further stirred for 12 hours at room temperature. After confirming that the starting material phosphite disappeared, the solvent was distilled off under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=4:1) to obtain the desired product (0.3g, melting point: 40 to 42°C, yield: 17%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

1.93, 2.08	(3H, s, s)
3.67, 3.77	(2H, s, s)
3.8	(3H, s)
7.1-7.67	(8H, m)

7.85 (1H, br)

PREPARATION EXAMPLE 23

Preparation of 4-chloro-4'-methylsulfonylmethyl-benzophenone (Compound No. VI-3)

4-bromomethyl-4'-chlorobenzophenone (3.1 g) and sodium methanesulfinate (1.5 g) were added to N,N-dimethylformamide (50 ml), and the mixture was stirred for 16 hours at room temperature. Water was added to the reaction mixture, and the mixture was extracted with ethyl acetate. The ethyl acetate layer was washed with water and dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residual solid was washed with n-hexane to obtain the desired product (2.8 g, melting point: 164 to 166°C, yield: 90%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

2.90	(3H, s)
4.47	(2H, s)
7.37-7.83	(8H, m)

PREPARATION EXAMPLE 24

Preparation of 4-chloro-4'-ethylsulfonylmethyl-benzophenone (Compound No. VI-6)

Sodium sulfite (24.5 g) and sodium hydrogen carbonate (33 g) were dissolved in water (200 ml). Ethane sulfonyl chloride (25 g) was dropwise added to this solution at room temperature in 30 minutes, and the mixture was stirred for one hour. This reaction mixture was

concentrated, and the residue was suspended in N,N-dimethylformamide (200 ml). Then, 4-bromomethyl-4'-chlorobenzophenone (10.0 g) was added thereto, and the mixture was stirred for 16 hours at room temperature. Water was added to the reaction mixture, and the mixture was extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residual solid was washed with n-hexane to obtain the desired product (7.5 g, melting point: 117 to 118°C, yield: 72%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

1.37	(3H, t)
2.93	(2H, q)
4.27	(2H, s)
7.20-7.83	(8H, m)

PREPARATION EXAMPLE 25

Preparation of 4-chloro-4'-ethylthiomethylbenzophenone (Compound No. VI-4)

4-chloro-4'-mercaptomethylbenzophenone (16.0 g), ethyl bromide (7.4 g) and potassium hydroxide (4.3 g) were added to methanol (250 ml), and the mixture was stirred for 30 minutes under reflux. The reaction mixture was cooled to room temperature and then concentrated. Water was added to the residue, and the mixture was extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over

anhydrous magnesium sulfate and concentrated. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=10:1) to obtain the desired product 14.0 g, melting point: 33 to 34°C, yield: 79%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

2.23	(3H, t)
2.45	(2H, s)
3.75	(2H, s)
7.10-7.90	(8H, m)

PREPARATION EXAMPLE 26

Preparation of 4-chloro-4'-difluoromethylthiomethyl-benzophenone (Compound No. VI-15)

4-chloro-4'-mercaptomethylbenzophenone (14.7 g) and potassium hydroxide (15 g) was added to a solvent comprising dioxane (100 ml) and water (100 ml). Difluoromethyl chloride was blown into this solution at 60°C until the starting material 4-chloro-4'-mercaptomethylbenzophenone disappeared. The reaction product was cooled to room temperature and then subjected to filtration. The organic layer as the filtrate was dried over anhydrous magnesium sulfate and concentrated. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=5:1) to obtain the desired product (6.4 g, melting point: 34 to 35°C, yield: 36%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

4.03	(2H, s)

6.69		(1H, t)
7.15-7.71		(8H, m)

PREPARATION EXAMPLE 27

Preparation of 4-chloro-4'-difluoromethylsulfonylmethyl-benzophenone (Compound No. VI-16)

4-chloro-4'-difluoromethylthiomethylbenzophenone (3.2 g) and m-chloroperbenzoic acid (5.3 g) were added to chloroform (150 ml). This suspension was stirred for 3 hours under reflux. The reaction mixture was concentrated, and water was added to the residue. The mixture was extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residual solid was washed with n-hexane to obtain the desired product (2.7 g, melting point: 1554 to 157°C, yield: 78%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

4.57		(2H, s)
6.41		(1H, t)
7.27-7.87		(8H, m)

PREPARATION EXAMPLE 28

Preparation of 4-chloro-4'-trifluoromethylthiomethyl-benzophenone (Compound No. VI-12)

4-chloro-4'-mercaptomethylbenzophenone (4.5 g) was dissolved in tetrahydrofuran (150 ml). 60% sodium hydride (0.8 g) was added to this solution, and the mixture was stirred for 30 minutes at room temperature.

Then, S-(trifluoromethyl)dibenzothiophenium-trifluoromethane sulfonate (6.4 g) was added thereto, and the mixture was further stirred for 30 minutes. The reaction mixture was concentrated, and water was added thereto. The mixture was extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=4:1) to obtain the desired product (2.0 g, melting point: 63 to 65°C, yield: 35%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

4.14	(2H, s)
7.30-7.77	(8H, m)

PREPARATION EXAMPLE 29

Preparation of 4-chloro-4'-(1,1,2,2-tetrafluoroethyl-thiomethylbenzophenone (Compound No. VI-20)

4-chloro-4'-mercaptomethylbenzophenone (5.0 g) and potassium tert-butoxide (0.9 g) were added to ethanol (150 ml). Perfluoroethylene (2.9 g) were blown thereinto at room temperature, and the mixture was then stirred for 16 hours. The reaction mixture was filtered and concentrated. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=4:1) to obtain the desired product 95.3g, melting point: 48 to 50°C, yield: 77%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

4.13		(2H, s)
5.77		(1H, tt)
7.23-7.73		(8H, m)

PREPARATION EXAMPLE 30

Preparation of 4-chloro-4'-methylthiomethylbenzophenone (Compound No. VI-1)

4-bromomethyl-4'-chlorobenzophenone (3.1 g) and 15% methylmercaptan-sodium aqueous solution (5.6 g) were added to methanol (150 ml), and the mixture was stirred for 30 minutes under reflux. The reaction mixture was concentrated and extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residual solid was washed with n-hexane to obtain the desired product (2.3 g, melting point: 59 to 61°C, yield: 83%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

2.00		(3H, s)
3.70		(2H, s)
7.13-7.74		(8H, m)

PREPARATION EXAMPLE 31

Preparation of 4-chloro-4'-methylsulfinylmethylbenzophenone (Compound No. VI-2)

4-chloro-4'-methylthiomethylbenzophenone (4.2 g) was added to methanol (150 ml). Sodium periodate (3.6 g) dissolved in water (20 ml), was added to this solution, and the mixture was stirred for 16 hours at room

temperature. The reaction mixture was concentrated, and water was added thereto. The mixture was extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residual solid was washed with n-hexane to obtain the desired product (4.1 g, melting point: 116 to 118°C, yield: 93%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

	2.50	(3H, s)
	4.00	(2H, s)
	7.30-7.80	(8H, m)

PREPARATION EXAMPLE 32

Preparation of 4-(3-bromopropyl)sulfonylmethyl-4'-chlorobenzophenone (Compound No. VI-11)

4-(3-bromopropyl)thiomethyl-4'-chlorobenzophenone (5.1 g) and a 31% hydrogen peroxide aqueous solution (6 g) was added to acetic acid (200 ml), and the mixture was stirred for one hour at 80°C and further for one hour under reflux. The reaction mixture was concentrated, and water was added thereto. The mixture was extracted with ethyl acetate. The ethyl acetate layer was washed with an aqueous potassium carbonate solution and water and dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residual solid was washed with n-hexane to obtain the desired product (5.0 g, melting pint: 105 to 107°C, yield: 91%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

2.17-2.60	(2H, m)
3.00-3.17	(2H, m)
3.53	(2H, t)
4.33	(2H, s)
7.23-7.87	(m, 8H)

PREPARATION EXAMPLE 33

Preparation of 4-chloro-4'-(1,1-dioxothiolan-2-yl)benzophenone (Compound No. VI-36)

4-(3-bromopropyl)sulfonylmethyl-4'-chlorobenzophenone (2.5 g) and 60% sodium hydride (0.3 g) were added to N,N-dimethylacetamide (70 ml), and the mixture was stirred for 16 hours at room temperature. Water was added to the reaction mixture, and the mixture was extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=1:1) to obtain the desired product as a slightly yellow viscous substance 91.0 g, yield: 50%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

2.67-2.73	(4H, m)
2.87-3.33	(2H, m)
4.02-4.37	(1H, m)
7.23-7.97	(8H, m)

PREPARATION EXAMPLE 34

Preparation of 4-chloro-4'-(2-trifluoromethylsulfonylpropyl)benzophenone (Compound No. VI-28)

4-chloro-4'-trifluoromethylsulfonylmethylbenzophenone (3.3 g) and 60% sodium hydride (0.8 g) were added to N,N-dimethylacetamide (150 ml), and the mixture was stirred for one hour at room temperature. Methyl iodide (0.8 g) was added to this solution, and the mixture was stirred for 16 hours at room temperature. Water was added to the reaction mixture, and the mixture was extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=4:1) to obtain the desired product (3.1 g, melting point 107 to 109°C, yield: 86%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

2.00	(6H, s)
7.20-7.70	(8H, m)

PREPARATION EXAMPLE 35

Preparation of 4-chloro-4'-thiocyanatomethylbenzophenone (Compound No. VI-35)

4-bromomethyl-4'-chlorobenzophenone (5.7 g) and sodium thiocyanate (5.5 g) were added to ethanol (50 ml), and the mixture was stirred for one hour at 60°C. The reaction mixture was concentrated, and water was added to

the residue. The mixture was extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residual solid was washed with a solvent mixture of n-hexane:ethyl acetate=10:1 to obtain the desired product (2.2 g, melting point: 129 to 131°C, yield:42%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

4.18	(2H, s)
7.23-7.87	(8H, m)

PREPARATION EXAMPLE 36

Preparation of diethyl 2-{4-(4-chlorobenzoyl)phenyl}-2-trifluoromethylthiomalonate (Compound No. VI-80)

60% sodium hydride (0.5 g) was dispersed in tetrahydrofuran (150 ml), and diethyl 2-{4-(4-chlorobenzoyl)phenyl} malonate (4.4 g) was dropwise added thereto at 0°C with stirring. After generation of hydrogen ceased, trifluoromethylsulphenyl chloride was blown thereinto at 0°C, and the mixture was then stirred for one hour at room temperature. The reaction mixture was concentrated, and water was added to the residue. The mixture was extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled off under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=8:1) to obtain the desired product (4.7 g,

n_D^{20}:1.5362, yield:87%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

1.3	(6H, t)
4.35	(4H, q)
7.4	(2H, d)
7.75	(2H, d)
7.8	(4H, s)

PREPARATION EXAMPLE 37

Preparation of 4-chloro-4'-trichloromethylthiomethylbenzophenone (Compound No. VI-81)

Methylbenzophenone 4-chloro-4'-thiocyanate (5.5 g) and triethylbenzylammonium chloride (0.5 g) were dispersed in chloroform (30 ml), and a 48% sodium hydroxide aqueous solution (4 ml) was added thereto at 40°C. Then, the mixture was stirred for 3 hours.

Cool water was added thereto, and the mixture was subjected to liquid separation. The organic layer was washed with water and dried over anhydrous magnesium sulfate. Then, chloroform was distilled off under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=9:1) to obtain the desired product (1.0 g, melting point: 103 to 105°C, yield: 13%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

4.45	(2H, s)
7.15-7.8	(8H, m)

PREPARATION EXAMPLE 38

Preparation of 4-chloro-4'-trifluoromethylsulfonylmethylbenzhydrol (Compound No. VIII-2)

4-chloro-4'-trifluoromethylsulfonylmethylbenzophenone (5.5 g) was dispersed in methanol (200 ml). Sodium borohydride was gradually added thereto at room temperature with stirring, and the mixture was further stirred overnight at room temperature. After completion of the reaction, methanol was distilled off under reduced pressure. The residue was extracted with ethyl acetate (250 ml). The extract was washed with water and dried over anhydrous magnesium sulfate. Then, ethyl acetate was distilled off under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=2:1) to obtain the desired product as a white powder (4.2 g, melting point: 113 to 115°C, yield: 77%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

	4.7	(2H, s)
	5.77	(2H, s)
	7.3	(4H, s)
	7.47	(4H, s)

PREPARATION EXAMPLE 39

Preparation of 4-chloro-4'-ethane sulfonylmethylbenzhydryl chloride (Compound No. VIII-5)

4-chloro-4'-ethane sulfonylmethylbenzhydrol (6.0 g),

thionyl chloride (5.4 g), toluene (200 ml) and a catalytic amount of N,N-dimethylformamide were mixed and gradually heated with stirring to a refluxing temperature. The mixture was stirred for 4 hours under reflux and then left to cool. Then, the solvent was distilled off under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=2:1) to obtain the desired product as a slight yellow viscous substance (4.6g, n_D^{20}: 1.6044, yield: 75%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

1.33	(3H, t)
2.88	(2H, q)
4.37	(2H, s)
6.05	(1H, s)
7.27	(4H, s)
7.35	(4H, s)

PREPARATION EXAMPLE 40

Preparation of (6-chloro-3-pyridyl)(4-trifluoromethylphenyl) ketone (Compound No. VII-3)

(6-chloro-3-pyridyl)(4-thicyanatemethylphenyl) ketone (5.0 g) was dissolved in tetrahydrofuran (300 ml), and trifluoromethyltrimethyl silane (5.0 g) was added thereto at room temperature. Then, the mixture was cooled to 5°C. Tetrabutylammonium fluoride (1.0 M tetrahydrofuran solution, 23 g) was gradually dropwise added thereto under cooling, and then, the mixture was stirred over

night. Tetrahydrofran was distilled off under reduced pressure, and the residue was extracted with ethyl acetate. The extract was washed with water and dried over anhydrous magnesium sulfate. Then, ethyl acetate was distilled off under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane:ethyl acetate=4:1) to obtain the desired product (2.0 g, n_D^{20}: 1.5820, yield: 35%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

4.15	(2H, s)
7.4	(3H, dd)
7.72	(2H, dd)
8.05	(2H, dd)
8.67	(2H, d)

PREPARATION EXAMPLE 41

Preparation of 4-chloro-4'-hydroxymethylbenzophenone-N'-methoxcarbonylhydrazone (Compound No. IX-1)

4-chloro-4'-hydroxymethylbenzophenone (0.5 g) and methyl-carbazate (0.4 g) were added to ethanol (60 ml) and acetic acid (5 ml), and the mixture was stirred for 2 hours under reflux. The reaction mixture was concentrated, and the residue was extracted with ethyl acetate. The ethyl acetate layer was washed with water and then dried over anhydrous magnesium sulfate. Ethyl acetate was distilled of under reduced pressure. The residue was purified by silica gel column chromatography (n-hexane: ethyl acetate=4:1) to obtain the desired

product (0.5 g, yield 83%).

^{1}H-NMR data (60MHz, $CDCl_3$ solvent, δ value)

2.17	(1H, br)
3.77	(3H, s)
4.63, 4.73	(2H, s, s)
6.97-7.63	(8H, m)
7.73	(1H, br)

When the compound of the present invention is to be used as the active component of a pesticide, it may be used by itself. However, it can be formulated into various formulations such as an emulsifiable concentrate, a suspension, a dust, a granule, a tablet, a wettable powder, a water-soluble concentrate, a solution, a flowable suspension, a water dispersible granule, an aerosol, a paste, an oil formulation, a concentrated emulsion in water in combination with various carriers, surfactants and other adjuvants which are commonly used for formulation as agricultural adjuvants. They are blended usually in such proportions that the active ingredient is from 0.1 to 90 parts by weight and the agricultural adjuvants are from 10 to 99.9 parts by weight.

The carriers to be used for such formulation may be classified into solid carriers and liquid carriers. The solid carriers include, for example, animal and plant powders such as starch, active carbon, soybean powder, wheat powder, wood powder, fish powder and powdered milk,

and mineral powders such as talc, kaolin, bentonite, calcium carbonate, zeolite, diatomaceous earth, fine silica powder, clay and alumina. The liquid carries include, for example, water, alcohols such as isopropyl alcohol and ethylene glycol, ketones such as cyclohexanone and methyl ethyl ketone, ethers such as dioxane and tetrahydrofuran, aliphatic hydrocarbons such as kerosene and light oil, aromatic hydrocarbons such as xylene, trimethylbenzene, tetramethylbenzene, methylnaphthalene and solvent naphtha, halogenated hydrocarbons such as chlorobenzene, acid amides such as dimethylacetamide, esters such as glycerin esters of fatty acids, nitriles such as acetonitrile, and sulfur-containing compounds such as dimethylsulfoxide.

The surfactants include, for example, metal salts of alkylbenzene sulfonic acids, metal salts of dinaphthylmethanedisulfonic acid, alcohol sulfuric acid esters, alkylarylsulfonates, lignin sulfonates, polyoxyethylene glycol ethers, polyoxyethylene alkyl aryl ethers and polyoxyethylene sorbitan monoalkylates.

Others adjuvants include, for example, an adhesive or thickener such as carboxymethylcellulose, gum arabic, sodium arignate, guar gum, tragacanth gum or polyvinyl alcohol, an antifoaming agent such as metal soap, a physical property-improving agent such as a fatty acid, an alkyl phosphate, silicone or paraffin, and a coloring agent.

When these formulations are to be practically used, they may be used as they are or as diluted with a diluting agent such as water to a predetermined concentration. Various formulations containing the compounds of the present invention or their diluted solutions may be applied by conventional methods i.e. application methods (such as spraying, misting, atomizing, dusting, granule application, paddy water application or seeding box treatment), soil treatment (such as mixing or drenching), surface application (such as painting, dressing or covering), dipping or poison bait. Further, the above active component may be fed as mixed in feeds to domestic animals, so that infestation or growth of pests, particularly injurious insects can be prevented by the excrements. Otherwise, it can also be applied by a so-called super high concentration low volume application method, whereby the active component may be contained up to 100%.

The pesticide of the present invention is applied usually in a concentration of the active ingredient of from 0.1 to 50,000 ppm, preferably from 1 to 10,000 ppm.

The concentration of the active ingredient can be suitably changed depending upon the type of the formulation, the method, the purpose, the season or the site of application and the state of infestation of pests. For example, in the case of aquatic pests, they can be controlled by applying a formulation having a

concentration within the above mentioned range to the infested site, and therefore, the range of the active ingredient in water is lower than the above range. The dose per unit area is usually from 0.1 to 5,000 g, preferably from 1 to 1,000 g, per 1 ha of the active compound. However, the dose is not limited to such a specific range.

The compound of the present invention is sufficiently effective when used alone. However, as a case requires, it may be used in combination or in admixture with fertilizers or other agricultural chemicals such as insecticides, acaricides, nematicides, fungicides, antivirus agents, attractants, herbicides or plant growth regulants, and further improved effects may sometimes be obtained by such combined use.

Typical examples of the insecticides, fungicides and acaricides which can be used in combination with the compound of the present invention, will be given below.

Organophosphorus compounds and carbamate insecticides: fenthion, fenitrothion, diazinon, chlorpyriphos, oxydeprofos, vamidothion, phenthoate (fentoat), dimethoate, formothion, malathion, trichlorphon, thiometon, phosmet, dichlorvos, acephate, EPBP (0-2,4-dichlorophenyl 0-ethylphenylphosphonothioate), methyl-parathion, oxydemeton-methyl, ethion, dioxabezofos, cyanophos (cyanofos), isoxathion, pyridafenthion, phosalone,

metidation, sulprophos (sulprofos), chlorfenvinphos, tetrachlorvinphos, dimethylvinphos, propahos, isofenphos, disulfoton, profenofos, pyraclofos, monocrotophos, azinphos-methyl, aldikarb, methomyl, thiodicarb, carbofuran, carbosulfan, benfuracarb, furathiocarb, propoxur, fenobcarb, metolcarb, isoprocarb, carbaryl (carbaril), pirimicarb, ethiofencarb, dichlophenthion, pirimiphos-methyl, quinalphos, chlorpyriphos-methyl, prothiophos, naled, bendiocarb, oxamyl, alanycarb, chlorethoxyfos, etc.

Pyrethroid insecticides: permethrin, cypermethrin, deltamethrin, fenvalerate, fenpropathrin, piretrine, allethrin, tetramethrin, resmethrin, dimethrin, proparthrin, phenothrin, prothrin, fluvalinate, cyfluthrin, cyhalothrin, flucythrinate, etofenprox, cycloprothrin, tralomethrin, silafluofen, tefluthrin, bifenthrin, acrinathrin, etc.

Acylurea type and other insecticides: diflubenzuron, chlorfluazuron, hexaflumuron, triflumuron, teflubenzuron, flufenoksuron, flucycloxuron, buprofezin, pyriproxyfen, lufenuron, cyromazine, methoprene, endosulphan, diafenthiuron, imidacloprid, fipronil, nicotin-sulfate, rotenone, metaldehyde, machine oil, fenoxycarb, cartap, thiocyclam, bensultap, tebufenozide, chlorphenapyr, emamectin-benzoate, acetamiprid, nitenpyram, pymetrozine, sodium oleate, rapeseed oil, etc.

Nematicides: phenamiphos, fosthiazate, ethoprophos,

methyl isothiocyanate, 1,3-dichloropropene, DCIP, etc.

Acaricides: chlororbenzilate, phenisobromolate, dicofol, amitraz, propargit, benzomate, hexythiazox, fenbutatin oxide, polynactins, quinomethionate, chlorfenson, tetradifon, avermectin, milbemectin, clofentezine, pyridaben, fenpyroximate, tebufenpyrad, pyrimidifen, fenothiocarb, dienochlor, etoxazole, halfenprox, etc.

Fungicides: thiophanate-methyl, benomil, carbendazol, thiabendazol, folpet, thiuram, diram, zineb, maneb, manzeb, polycarbamate, iprobenfos, edifenphos, fthalide, probenazole, isoprothiolane, chlorothalonil, captan, polyoxin, blasticidin-S, kasugamycin, streptomycin, validamycin, tricyclazole, pyroquilone, phenazine oxide, mepronil, flutolanil, pencycuron, iprodione, hymexazol, metalaxyl, triflumizole, triforine, triadimefone, bitertanol, fenarimol, propikonazol, cymoxanil, prochloraze, pefurazoate, hexaconazole, myclobutanil, diclomezine, tecloftalam, propineb, dithianon, phosethyl, vinclozolin, procymidone, oxadixyl, guazatine, propamocarb-hydrochloride, fluazinam, oxolinic acid, hydroxyisoxazole, imibenconazole, difenoconazole, mepanipyrim.

The compounds of the present invention exhibit excellent pesticidal activities against pests such as hemipteran insects, lepidopteran insects, coleopteran insects, dipteran insects, hymenopteran insects,

orthopteran insects, isopteran insects, thysanopteran insects, mites and plant-parastic nematodes. The following pests may be mentioned as such pests.

Hemipteran insects: bugs (HETEROPTERA) such as bean bug (Riptortus clavatus), southern green stink bug (Nezara viridula), lygus bugs (Lygus sp.), hairy chinch bug (Blissus leucopterus) and pear lace bug (Stephanitis nashi); leafhoppers (Circulifer sp.) such as green rice leafhopper (Nephotettix cincticeps) and leafhoppers (Empoasca sp., Erythroneura sp., Circulifer sp.); planthoppers (Delphacidae) such as brown rice planthopper (Nilaparvata lugens), whitebacked planthopper (Sogatella furcifera) and small brown planthopper (Laodelphax striatellus); jumping plantlice (Psyllidae) such as Psyllids (Psylla sp.); whiteflies (Aleyrodidae) such as sweetpotato whitefly (Bemisia tabaci) and greenhouse whitefly (Trialeurodes vaporariorum); aphides (Aphididae) such as grapeleaf louse (Viteus vitifolii), green peach aphid (Myzus persicae), green apple aphid (Aphis pomi), cotton aphid (Aphis gossypii), Aphis fabae, turnip aphid (Rhopalosiphum psedobrassicas), glasshouse-potato aphid (Aulacorthum solani) and greenbug (Schizaphis graminum); mealy bugs or scales such as comstock mealybug (Pseudococcus comstocki), red wax scale (Ceroplastes rubens), San Jose scale (Comstockaspis perniciosa) and arrowhead scale (Unaspis yanonensis).

Lepidopteran insects: tortricids (Tortricidae) such

as oriental tea tortrix (Homona magnanima), summer fruit tortrix (Adoxophyes orana), torticids (Sparganothis pilleriana), oriental fruit moth (Grapholitha molesta), soybean pod borer (Leguminivora glycinivorella), codling moth (Laspeyresia pomonella), tortricids (Eucosma sp.) and grape berry moth (Lobesia botrana); Cochylidae such as grape cochylid (Eupoecillia ambiguella); bagworm moths (Psychidae) such as Bambalina sp.; tineids (Tineidae) such as European grain moth (Nemapogon granellus) and casemaking clothes moth (Tinea translucens); lyonetiid moths (Lyonetiidae) such as Lyonetia prunifoliella; leafblotch miners such as apple leafminer (Phyllonorycter ringoniella); Phyllocnistidae such as citrus leafminer (Phyllocnistis citrella); yponomeutids such as diamondback moth (Plutella xylostella) and yponomeutid moths (Prays citri); clearwing moths (Synanthedon sp.) such as grape clearwing moth (Paranthrene regalis) and Synanthedon sp.; gelechiid moths (Gelechiidae) such as pink bollworm (Pectinophora gossypiella), potato tuberworm (Phthorimaea operculella) and Stomopteryx sp.; Carposinidae such as peach fruit moth (Carposina niponensis); slug caterpillarmoths such as oriental moth (Monema flavescens); pyralid moths such as rice stem borer (Chilo suppressalis), rice leafroller (Cnaphalocrocis medinalis), European corn borer (Ostrinia nubilalis), oriental corn borer (Ostrinia furnacalis), cabbage webworm (Hellula undalis), greater wax moth

(Galleria mellonella), lesser cornstalk borer (Elasmopalpus lignosellus) and beet webworm (Loxostege sticticalis); whites such as common cabbage worm (Pieris rapae); geometrid moths such as mugwort looper (Ascotis selenaria); tent caterpillar moths such as tent caterpillar (Malacosoma neustria); sphinx moths such as tobacco hornworm (Manduca sexta); tussock moths such as tea tussock moth (Euproctis pseudoconspersa) and gypsy moth (Lymantria dispar); tiger moths such as fall webworm (Hyphantria cunea); owlet moths such as tobacco budworm (Heliothis virescens), bollworm (Helicoverpa zea), beet armyworm (Spodoptera exigua), cotton bollworm (Helicoverpa armigera), common cutworm (Spodoptera litura), cabbage armyworm (Mamestra brassicae), black cutworm (Agrotis ipsiron), rice armyworm (Pseudaletia separata) and cabbage looper (Trichoplusia ni).

Coleopteran insects: chafers such as cupreous chafer (Anomala cuprea), Japanese beetle (Popillia japonica), soybean beetle (Anomala rufocuprea) and Eutheola rugiceps; click beetles (Conodeus sp.) such as wireworm (Agriotes sp.) and Conodeus sp.; ladybird beetles such as twenty-eight-spotted ladybird (Epilachna vigintioctopunctata) and Mexican bean beetle (Epilachna varivestis); darkling beetles such as red-brown rice-flour beetle (Tribolium castaneum); longicorn beetles such as whitespotted longicorn beetle (Anoplophora malasiaca) and Japanese pine sawyer (Monochamus

alternatus); seed beetles such as bean weevil (Acanthoscelides obtectus) and adzuki bean weevil (Callosobruchus chinensis); leaf beetles such as colorado potato beetle (Leptinotarsa decemlineata), corn rootworm (Diabrotica sp.), rice leaf beetle (Oulema oryzae), beet flea beetle (Chaetocnema concinna), mustard beetle (Phaedon cochlearias), cereal leaf beetle (Oulema melanopus) and Dicladispa armigera; Apionidae such as Apion godmani; weevils such as rice water weevil (Lissorhoptrus oryzophilus) and cotton boll weevil (Anthonomus grandis); Rhynchophoridae such as maize weevil (Sitophilus zeamais); bark beetles; skin beetles; drugstore beetles.

Dipteran insects: rice crane fly (Tipra ano), rice midge (Tanytarsus oryzae), Orseolia oryzae, Ceratitis capitata, rice leafminer (Hydrellia griseola), cherry drosophila (Drosophila suzukii), frit fly (Oscinella frit), rice stem maggot (Chlorops oryzae), French bean miner (Ophiomyia phaseoli), legume leafminer (Liriomyza trifolii), beet leafminer (Pegomya hyoscyami), seedcorn maggot (Hylemia platura), sorghum fly (Atherigona soccata), muscid fly (Musca domestica), Gastrophilus sp., stomoxiid flies (Stomoxys sp.), Aedes aegypti, Culex pipiens, Anopheles sinensis and Culex tritaeniorhynchus.

Hymenopteran insects: stem sawflies (Cephus sp.); eurytomids (Harmolita sp.); cabbage sawfly (Athalia sp.), hornets (Vespa sp.) and fire ants.

Orthopteran insects: German cockroach (Blatella germanica); American cockroach (Periplaneta americana); mole crichet (Gryllotalpa africana); Asiatic locust (Locusta migratoria migratoriodes); and Melanoplus sanquinipes.

Termites insects: termites (Reticulitermes speratus) and formosan subterranean termite (Coptotermes formosanus).

Thrips insects: yellow tea thrips (Scirtothrips dorsalis); thrips (Thrips palmi); greenhouse thrips (Heliothrips haemorrholidalis); western flower thrips (Frankliniella occidentalis) and rice aculeated thrips (Haplothrips aculeatus).

Mites: twospotted spider mite (Tetranychus urticae); Kanzawa spider mite (Tetranychus kanzawai); citrus red mite (Panonychus citri); European red mite (Panonychus ulmi), yellow spider mite (Eotetranychus carpini); Texas citrus mite (Eotetranychus banksi); citrus rust mite (Phyllocoptruta oleivora); broad mite (Polyphagotarsonemus latus); false spider mites (Brevipalpus sp.); bulb mite (Rhizoglyphus robini) and mold mite (Tyrophagus putrescentiae).

Plant-parasitic nematodes: southern root-knot nematode (Meloidogyne incognita); root-lesion nematode (Pratylenchus sp.); soybean cyst nematode (Heterodera glycines); rice white-tip nematode (Aphelenchoides besseyi) and pine wood nematode (Bursaphelenchus

xylophilus).

Other pests and parasites: Gastropoda such as apple snails (Pomacea canaliculata); slugs (Incilaria sp.) and giant African snail (Achatina fulica); pillbugs (Isopoda) such as sow bug and centipede; booklice (Liposcelis sp.); oriental siverfish (Ctenolepisma sp.); Pulex sp.; Trichodectes sp.; Cimex sp.; aminal-parasitic mites such as Boophilus microplus and aemaphysalis longicornis and Epidermoptidae.

Further, the compounds of the present invention are effective also against pests which show resistance to organophosphorus compounds, carbamate compounds, synthetic pyrethroid compounds, acylurea compounds or conventional insecticides.

Thus, compounds of the present invention exhibit excellent pesticidal effects against a wide range of pests including hemipteran insects, lepidopteran insects, coleopteran insects, dipteran insects, hymenopteran insects, orthopteran insects, isopteran insects, thysanopteran insects, mites and plant-parastic nematodes, and they are also capable of controlling pests which acquired resistance to conventional pesticides.

Now, formulation methods will be described in detail with reference to typical Formulation Examples. However, it should be understood that the types and the proportions of the compounds and the adjuvants are not restricted by these specific Examples and may be varied

within wide ranges. In the following examples, "%" means "% by weight".

FORMULATION EXAMPLE 1: Emulsifiable concentrate

30% of compound (I-22), 20% of cyclohaxanone, 11% of polyoxyethylene alkylaryl ether, 4% of calcium alkylbenzene sulfonate and 35% of methylnaphthalene were uniformly dissolved to obtain an emulsifiable concentrate.

FORMULATION EXAMPLE 2: Wettable powder

10% of compound (I-22), 0.5% of a sodium salt of a naphthalene sulfonic acid/formalin condensation product, 0.5% of polyoxyethylene alkylaryl ether, 24% of diatomaceous earth and 65% of clay were uniformly mixed and pulverized to obtain a wettable powder.

FORMULATION EXAMPLE 3: Dust

2% of compound (I-22), 5% of diatomaceous earth and 93% of clay were uniformly mixed and pulverized to obtain a dust.

FORMULATION EXAMPLE 4: Granule

5% of compound (I-22), 2% of sodium lauryl alcohol sulfate, 5% of sodium lignin sulfonate, 2% of carboxymethylcellulose and 86% of clay were uniformly mixed and pulverized. 100 parts by weight of this mixture was kneaded with 20 parts by weight of water and formed into granules of from 14 to 32 mesh by an extrusion-type granulator, followed by drying to obtain a granule formulation.

Now, the effects of the pesticides containing the compounds of the present invention as active ingredients will be described with reference to Test Examples. Comparative Compounds a and b are compounds disclosed in Example 165 and Example 6 in Japanese Unexamined Patent Publication No. 122261/1979; Comparative Compound c is a compound disclosed in Example 88 in Japanese Unexamined Patent Publication No. 45452/1981; and Comparative Compound d is a compound disclosed in Example 6 in U.S. Patent 3,732,3067. These Comparative Compounds were formulated and used in the same manner as the compounds of the present invention.

Comparative Compound a: 4-chloro4'-isopropylthiobenzophenone-N'-ethoxycarbonylhydrazone

Comparative Compound b: 4-chloro-4'-propylsulfonylbenzophenone-N'-propionylhydrazone

Comparative Compound c: 4-chloro-4'-methylsulfinylbenzophenone-N'-ethoxycabonylhydrazone

Comparative Compound d: 4-trifluoromethylbenzophenonehydrazone

Test Example 1 Insecticidal test for diamondback moth

The wettable powder prepared according to Formulation Example 2 was diluted with water so that the concentration of the active ingredient was 500 ppm. Cabbage leaves were immersed in the resulting diluted solution, dried in air and then placed in a polyvinyl chloride cup. Ten larvae of diamondback moth were

released in the cup, and thereafter a cover was placed thereon. Then, the cup was placed in a thermostatic chamber of 25°C for 6 days, and the number of insects died was counted to calculate the mortality (%) according to a calculation formula (A). The results are shown in Table 36. The test was carried out in two series.

$$\text{Mortality (\%)} = \frac{\text{Number of insects died}}{\text{Number of insects released}} \times 100 \quad \text{(A)}$$

Table 36

Compound No.	Mortality
I-1	100
I-2	100
I-10	100
I-11	100
I-12	100
I-13	100
I-14	100
I-15	100
I-16	100
I-17	100
I-18	100
I-19	100
I-20	100
I-21	100
I-22	100
I-23	100
I-24	100
I-25	100
I-26	100
I-31	100
I-32	100
I-34	100
I-35	100
I-40	100
I-41	100
I-42	100
I-43	100
I-44	100
I-45	100
I-46	100
I-47	100

Table 36 (continued)

Compound No.	Mortality
I-48	100
I-50	100
I-51	100
I-52	100
I-53	100
I-54	100
I-55	100
I-56	100
I-57	100
I-58	100
I-59	100
I-60	100
I-61	100
I-62	100
I-63	100
I-64	100
I-65	100
I-66	100
I-67	100
I-68	100
I-69	100
I-70	100
I-71	100
I-72	100
I-73	100
I-74	100
I-75	100
I-76	100
I-77	100
I-78	100
I-81	100

Table 36 (continued)

Compound No.	Mortality
I-83	100
I-84	100
I-88	100
I-89	100
I-90	100
I-91	100
I-92	100
I-93	100
I-94	100
I-95	100
I-96	100
I-97	100
I-98	100
I-99	100
I-100	100
I-101	100
I-102	100
I-106	100
I-114	100
I-115	100
I-116	100
I-117	100
I-118	100
I-119	100
I-121	100
I-122	100
I-124	100
I-125	100
I-128	100
I-129	100
I-130	100

Table 36 (continued)

Compound No.	Mortality
I-132	100
I-133	100
I-134	100
I-135	100
I-136	100
I-137	100
I-138	100
I-140	100
I-141	100
I-148	100
I-149	100
I-150	100
I-151	100
I-153	100
I-154	100
I-155	100
I-156	100
I-157	100
I-158	100
I-159	100
I-160	100
I-161	100
I-162	100
I-163	100
I-164	100
I-165	100
I-166	100
I-168	100
I-169	100
I-171	100
I-173	100

Table 36 (continued)

Compound No.	Mortality
I-174	100
I-175	100
I-176	100
I-177	100
I-178	100
I-179	100
I-181	100
I-182	100
I-183	100
I-184	100
I-185	100
I-186	100
I-187	100
I-188	100
I-189	100
I-190	100
I-191	100
I-192	100
I-193	100
I-194	100
I-195	100
I-196	100
I-197	100
I-206	100
I-207	100
I-209	100
I-210	100
I-211	100
I-212	100
I-213	100
I-214	100

Table 36 (continued)

Compound No.	Mortality
I-216	100
I-217	100
I-218	100
I-220	100
I-221	100
I-222	100
I-223	100
I-224	100
I-225	100
I-226	100
I-238	100
I-247	100
I-250	100
I-251	100
I-252	100
I-253	100
I-254	100
I-255	100
I-256	100
I-257	100
I-258	100
I-259	100
I-260	100
I-261	100
I-263	100
I-264	100
I-265	100
I-266	100
I-268	100
I-269	100
I-274	100

Table 36 (continued)

Compound No.	Mortality
I-277	100
I-278	100
I-280	100
I-281	100
I-282	100
I-283	100
I-284	100
I-285	100
I-286	100
I-290	100
I-291	100
I-292	100
I-293	100
I-294	100
I-295	100
I-296	100
I-297	100
I-298	100
I-299	100
I-300	100
I-301	100
I-302	100
I-303	100
I-305	100
I-306	100
I-307	100
I-308	100
I-309	100
I-310	100
I-311	100
I-312	100

Table 36 (continued)

Compound No.	Mortality
I-313	100
I-314	100
I-315	100
I-316	100
I-317	100
I-318	100
I-319	100
I-320	100
I-321	100
I-323	100
I-324	100
I-326	100
I-328	100
I-329	100
I-330	100
I-331	100
I-332	100
I-333	100
I-334	100
I-335	100
I-336	100
I-337	100
I-338	100
I-339	100
I-340	100
I-341	100
I-342	100
I-343	100
I-344	100
I-345	100
I-346	100

Table 36 (continued)

Compound No.	Mortality
I-347	100
I-348	100
I-349	100
I-350	100
I-351	100
I-352	100
I-353	100
I-354	100
I-355	100
I-356	100
I-357	100
I-358	100
I-359	100
I-360	100
I-361	100
I-362	100
I-363	100
I-364	100
I-365	100
I-366	100
I-367	100
I-368	100
I-370	100
I-371	100
I-372	100
I-373	100
I-374	100
I-376	100
I-377	100
I-379	100
I-380	100

Table 36 (continued)

Compound No.	Mortality
I-381	100
I-382	100
I-383	100
I-384	100
I-385	100
I-386	100
I-387	100
I-388	100
I-389	100
I-390	100
I-391	100
I-392	100
I-393	100
I-394	100
I-395	100
I-396	100
I-397	100
I-398	100
I-399	100
I-400	100
I-401	100
I-402	100
I-403	100
I-404	100
I-405	100
I-407	100
I-408	100
I-409	100
I-410	100
I-411	100
I-412	100

Table 36 (continued)

Compound No.	Mortality
I-414	100
I-415	100
I-416	100
I-417	100
I-418	100
I-419	100
I-420	100
I-421	100
I-423	100
I-424	100
I-425	100
I-426	100
I-428	100
I-429	100
I-431	100
I-432	100
I-435	100
I-436	100
I-437	100
I-438	100
I-439	100
I-440	100
I-441	100
I-442	100
I-443	100
I-444	100
I-445	100
I-447	100
I-448	100
I-449	100
I-450	100

Table 36 (continued)

Compound No.	Mortality
I-451	100
I-452	100
I-453	100
I-455	100
I-456	100
I-457	100
I-458	100
I-459	100
I-460	100
I-461	100
I-462	100
I-463	100
I-464	100
I-466	100
I-467	100
I-468	100
I-469	100
I-470	100
I-471	100
I-472	100
I-473	100
I-474	100
I-475	100
I-476	100
I-477	100
I-478	100
I-480	100
I-481	100
I-482	100
I-483	100
I-484	100

Table 36 (continued)

Compound No.	Mortality
I-485	100
I-486	100
I-487	100
I-488	100
I-489	100
I-490	100
I-491	100
I-492	100
I-494	100
I-495	100
I-496	100
I-497	100
I-498	100
I-499	100
I-500	100
I-501	100
I-502	100
I-504	100
I-509	100
I-510	100
I-516	100
I-517	100
I-518	100
I-519	100
I-520	100
I-521	100
I-522	100
I-523	100
I-524	100
I-525	100
I-526	100

Table 36 (continued)

Compound No.	Mortality
I-527	100
I-528	100
I-529	100
I-530	100
I-531	100
I-532	100
I-533	100
I-534	100
I-535	100
I-536	100
I-537	100
I-538	100
I-539	100
I-540	100
II-5	100
II-6	100
II-8	100
II-10	100
II-11	100
II-12	100
II-13	100
II-14	100
II-15	100
II-16	100
II-22	100
II-23	100
II-24	100
II-29	100
II-30	100
II-32	100
II-35	100

Table 36 (continued)

Compound No.	Mortality
II-36	100
II-37	100
II-38	100
II-39	100
II-40	100
II-41	100
II-42	100
II-43	100
II-44	100
II-45	100
II-46	100
II-47	100
II-48	100
II-49	100
II-50	100
II-51	100
II-52	100
III-1	100
III-2	100
III-3	100
III-4	100
III-5	100
III-6	100
III-7	100
III-8	100
III-9	100
III-10	100
III-11	100
IV-1	100

Table 36 (continued)

Compound No.	Mortality
V-1	100
V-2	100
V-3	100
V-4	100
V-5	100
V-6	100
V-7	100
V-8	100
V-9	100
V-10	100
V-11	100
V-12	100
V-13	100
V-14	100
V-15	100
V-16	100
V-17	100
V-18	100
V-19	100
VI-2	100
VI-15	100
VI-17	100
VI-22	100
Comparative d	0

Test Example 2 Insecticidal test for brown rice planthopper

The wettable powder prepared according to Formulation Example 2 was diluted with water so that the concentration of the active ingredient was 500 ppm. In the resulting diluted wettable powder were immersed rice stems and leaves, which were then dried in air and placed in a test tube. In the test tube were released 5 larvae of brown rice planthopper, and then the opening of the test tube was plugged with absorbent wadding. Thereafter, the test tube was placed in a thermostatic chamber of 25°C for 6 days, and then the number of insects died was counted to calculate the mortality (%) according to the calculation formula (A). The test was carried out in two series. The results are shown in Table 37.

Table 37

Compound No.	Mortality
I-10	100
I-11	100
I-12	100
I-13	100
I-14	100
I-15	100
I-32	100
I-41	100
I-45	100
I-46	100
I-47	100
I-50	100
I-51	100
I-52	100
I-53	100
I-55	100
I-61	100
I-62	100
I-64	100
I-65	100
I-66	100
I-67	100
I-68	100
I-69	100
I-70	100
I-71	100
I-72	100
I-73	100
I-74	100
I-75	100
I-77	100

Table 37 (continued)

Compound No.	Mortality
I-81	100
I-83	100
I-88	100
I-89	100
I-90	100
I-91	100
I-92	100
I-93	100
I-96	100
I-97	100
I-98	100
I-99	100
I-100	100
I-101	100
I-102	100
I-106	100
I-114	100
I-115	100
I-116	100
I-117	100
I-118	100
I-121	100
I-122	100
I-132	100
I-135	100
I-136	100
I-150	100
I-151	100
I-157	100
I-158	100
I-163	100

Table 37 (continued)

Compound No.	Mortality
I-164	100
I-165	100
I-173	100
I-176	100
I-177	100
I-179	100
I-180	100
I-183	100
I-184	100
I-185	100
I-186	100
I-187	100
I-188	100
I-190	100
I-194	100
I-211	100
I-212	100
I-213	100
I-214	100
I-216	100
I-217	100
I-218	100
I-220	100
I-221	100
I-222	100
I-224	100
I-225	100
I-249	100
I-250	100
I-252	100
I-253	100

Table 37 (continued)

Compound No.	Mortality
I-254	100
I-255	100
I-256	100
I-257	100
I-259	100
I-260	100
I-264	100
I-285	100
I-290	100
I-291	100
I-293	100
I-294	100
I-295	100
I-296	100
I-297	100
I-298	100
I-299	100
I-300	100
I-301	100
I-302	100
I-304	100
I-305	100
I-306	100
I-308	100
I-313	100
I-314	100
I-317	100
I-318	100
I-319	100
I-320	100
I-328	100

Table 37 (continued)

Compound No.	Mortality
I-329	100
I-330	100
I-333	100
I-334	100
I-335	100
I-336	100
I-337	100
I-338	100
I-339	100
I-340	100
I-341	100
I-344	100
I-346	100
I-347	100
I-348	100
I-349	100
I-350	100
I-351	100
I-352	100
I-353	100
I-354	100
I-355	100
I-356	100
I-357	100
I-358	100
I-359	100
I-360	100
I-361	100
I-362	100
I-363	100
I-364	100

Table 37 (continued)

Compound No.	Mortality
I-366	100
I-367	100
I-368	100
I-388	100
I-390	100
I-392	100
I-394	100
I-395	100
I-399	100
I-402	100
I-403	100
I-414	100
I-415	100
I-416	100
I-418	100
I-436	100
I-437	100
I-438	100
I-439	100
I-440	100
I-444	100
I-445	100
I-446	100
I-447	100
I-450	100
I-451	100
I-452	100
I-453	100
I-454	100
I-466	100
I-467	100

Table 37 (continued)

Compound No.	Mortality
I-468	100
I-470	100
I-472	100
I-473	100
I-474	100
I-475	100
I-480	100
I-481	100
I-482	100
I-483	100
I-484	100
I-485	100
I-486	100
I-487	100
I-488	100
I-489	100
I-490	100
I-491	100
I-494	100
I-496	100
I-497	100
I-502	100
I-510	100
I-516	100
I-517	100
I-520	100
I-521	100
I-522	100
I-524	100
I-525	100
I-526	100

Table 37 (continued)

Compound No.	Mortality
I-527	100
I-528	100
I-529	100
I-530	100
I-531	100
I-532	100
I-533	100
I-534	100
I-535	100
I-536	100
I-537	100
I-538	100
I-539	100
I-540	100
II-10	100
II-12	100
II-13	100
II-14	100
II-23	100
II-29	100
II-30	100
II-36	100
II-37	100
II-51	100
II-52	100
III-2	100
III-4	100
III-6	100
III-7	100
III-8	100
III-9	100

Table 37 (continued)

Compound No.	Mortality
III-10	100
III-11	100
IV-1	100
V-1	100
V-3	100
V-4	100
V-5	100
V-6	100
V-7	100
V-9	100
V-19	100
VI-22	100
VI-43	100
VI-80	100
Comparative a	10
Comparative b	20
Comparative c	20

Test Example 3 Insecticidal test for adzuki bean weevil

The wettable powder prepared according to Formulation Example 2 was diluted with water to a concentration of 100 ppm. 0.75 ml of this diluted solution was dropped on a filter paper having diameter of 6 cm placed in a polyvinyl chloride cup having a capacity of 60 ml. Five female adults of adzuki bean weevil were released in the cup, and a cover was placed thereon. Then, the cup was placed in a thermostatic chamber of 25°C for 4 days, and the number of insects died was counted to calculate the mortality (%) according to calculation formula (A). The test was carried out in two series. The results are shown in Table 38.

Table 38

Compound No.	Mortality
I-11	100
I-44	100
I-45	100
I-67	100
I-69	100
I-88	100
I-89	100
I-90	100
I-94	100
I-96	100
I-97	100
I-99	100
I-102	100
I-106	100
I-114	100
I-115	100
I-116	100
I-118	100
I-140	100
I-141	100
I-149	100
I-150	100
I-151	100
I-153	100
I-155	100
I-156	100
I-157	100
I-160	100
I-161	100
I-162	100
I-165	100

Table 38 (continued)

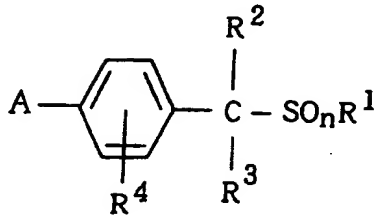
Compound No.	Mortality
I-173	100
I-174	100
I-175	100
I-176	100
I-177	100
I-178	100
I-179	100
I-180	100
I-183	100
I-185	100
I-186	100
I-187	100
I-188	100
I-189	100
I-191	100
I-192	100
I-194	100
I-195	100
I-196	100
I-197	100
I-206	100
I-218	100
I-222	100
I-253	100
I-254	100
I-257	100
I-258	100
I-263	100
II-5	100
II-6	100
II-10	100

Table 38 (continued)

Compound No.	Mortality
II-11	100
II-12	100
II-13	100
II-15	100
II-16	100
Comparative b	0
Comparative d	0

CLAIMS

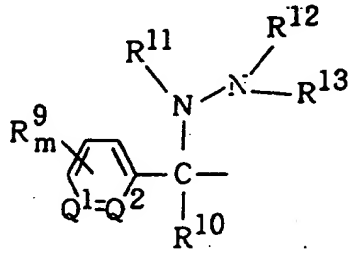
1. A benzylsulfide derivative of the formula (I) or its salt:



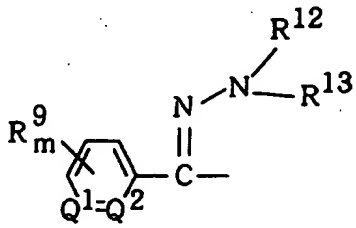
(I)

wherein R^1 is a C_{1-6} alkyl group, a C_{1-4} cyanoalkyl group, a C_{1-4} hydroxyalkyl group, a C_{3-6} cycloalkyl group, a C_{1-6} haloalkyl group, a C_{2-4} alkenyl group, a C_{2-4} alkynyl group, a phenyl group (which may be substituted by a halogen atom or a C_{1-4} alkyl group), a cyano group, a benzyl group (which may be substituted by a halogen atom), a thiazolyl group, a C_{1-4} alkylcarbamoyl group or a group of the formula $-N(R^5)R^6$; each of R^2 and R^3 which are independent of each other, is a hydrogen atom, a halogen atom, a cyano group, a C_{1-4} alkyl group, a C_{1-3} haloalkyl group, a C_{1-4} alkylthio group, a C_{1-4} alkylcarbonyl group, a carboxyl group, or a C_{1-4} alkoxycarbonyl group; or R^2 and R^3 may form a 3- to 6-membered ring together with the carbon atom to which they are bonded; or R^1 and R^2 may form a 3- to 8-membered ring having one or more hetero atoms, together with the sulfur and carbon atoms to which they are respectively bonded; R^4 is a hydrogen atom, a halogen atom, a C_{1-4} alkyl group, a C_{1-4} haloalkyl group, a C_{1-4} alkoxy group or a C_{1-4} haloalkoxy group; each of R^5 and R^6 which are

independent of each other, is a hydrogen atom, a C_{1-6} alkyl group or a C_{1-4} haloalkyl group; or R^5 and R^6 may together form a group of the formula $=CR^7R^8$; or R^5 and R^6 may form a 4- to 8-membered ring having one or more hetero atoms, together with the nitrogen atom to which they are bonded; R^7 is a hydrogen atom, a C_{1-3} alkyl group or a C_{1-3} alkylthio group; R^8 is a C_{1-3} alkylthio group or a C_{1-3} alkylamino group; or R^7 and R^8 may form a saturated or unsaturated 4- to 8-membered ring together with the carbon atom to which they are bonded; A is a hydrazinoaralkyl or hydrazonoaralkyl group of the formula (A1) or (A2);



[A1]



[A2]

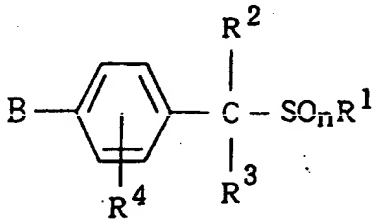
R^9 is a hydrogen atom, a halogen atom, a nitro group, a cyano group, a C_{1-4} alkyl group, a C_{1-4} haloalkyl group, a C_{1-4} alkoxy group, a C_{1-4} haloalkoxy group, a C_{1-4} alkylthio group, a C_{1-4} haloalkylthio group, a C_{1-4} alkylsulfonyl group, a C_{2-4} alkylsulfonylmethyl group, a C_{1-4} haloalkylsulfonyloxy group, a phenyl group (which may be substituted by a halogen atom) or a phenoxy group (which may be substituted by a halogen atom); or two R^9 may together form a 5- or 6-membered ring; R^{10} is a

hydrogen atom or a C_{1-4} alkyl group; each of R^{11}, R^{12} and R^{13} which are independent of one another, is a hydrogen atom, a cyano group, a C_{1-6} alkyl group, a C_{1-4} haloalkyl group, a C_{2-10} alkoxyalkyl group, a C_{3-8} alkoxyalkoxyalkyl group, a C_{2-6} alkylthioalkyl group, a C_{2-6} alkenyl group, a C_{2-4} alkynyl group, a C_{1-4} cyanoalkyl group, a benzyl group (which may be substituted by a halogen atom, a C_{1-4} haloalkyl group or a C_{1-4} alkyl group), a group of the formula $-COR^{14}$, a group of the formula $-CSR^{14}$, a group of the formula $-COOR^{15}$, a group of the formula $-COSR^{15}$, a group of the formula $-CON(R^{16})R^{17}$, a group of the formula $-CSN(R^{16})R^{17}$, a group of the formula $-SN(R^{18})R^{19}$, a group of the formula $-SO_2R^{20}$ or a group of the formula $-C(R^{21})=CHR^{22}$; or R^{12} and R^{13} may together form a group of the formula $=CR^{23}R^{24}$; or R^{12} and R^{13} may form a 4- to 8-membered ring having one or more hetero atoms, together with the nitrogen atom to which they are bonded; R^{14} is a hydrogen atom, a C_{1-20} alkyl group, a C_{1-8} haloalkyl group, a C_{2-12} alkoxyalkyl group, a C_{2-10} haloalkoxyalkyl group, a C_{3-16} alkoxyalkoxyalkyl group, a C_{4-22} alkoxyalkoxyalkoxyalkyl group, a C_{2-6} alkylthioalkyl group, a C_{3-6} cycloalkyl group, a C_{1-6} hydroxyalkyl group, a C_{1-6} aminoalkyl group, a C_{1-6} amidoalkyl group, a C_{1-8} cyanoalkyl group, a C_{3-12} alkoxycarbonylalkyl group, a C_{2-6} alkenyl group, a C_{2-4} alkynyl group, a phenyl group (which may be substituted by a halogen atom, a nitro group, a C_{1-4} alkyl group, a C_{1-4} haloalkyl group, a

phenoxy group or a C_{1-4} alkoxy group), a naphthyl group (which may be substituted by a halogen atom or a C_{1-4} alkyl group) or a hetero aromatic ring group (which may be substituted by a halogen atom or a C_{1-4} alkyl group); R^{15} is a C_{1-20} alkyl group, a C_{2-8} haloalkyl group, a C_{2-12} alkoxyalkyl group, a C_{2-6} alkenyl group, a C_{2-4} alkynyl group, a benzyl group (which may be substituted by a halogen atom, a C_{1-4} alkoxy group or a C_{1-4} alkyl group) or a phenyl group (which may be substituted by a halogen atom); R^{16} is a hydrogen atom or a C_{1-4} alkyl group; R^{17} is a hydrogen atom, a C_{1-6} alkyl group or a phenyl group (which may be substituted by a halogen atom, a C_{1-4} haloalkoxy group or a C_{1-4} alkyl group); each of R^{18} and R^{19} which are independent of each other, is a C_{1-4} alkyl group (which may be substituted by a C_{1-4} alkoxycarbonyl group), or a C_{2-5} alkoxyalkyl group; or R^{18} and R^{19} may form a 5- or 6-membered ring together with the nitrogen atom to which they are bonded; R^{20} is a C_{1-4} alkyl group, a C_{1-4} haloalkyl group or a C_{2-4} dialkylamino group; R^{21} is a hydrogen atom or a C_{1-6} alkyl group; R^{22} is a C_{2-4} acyl group or a C_{2-6} alkoxycarbonyl group; each of R^{23} and R^{24} which are independent of each other, is a hydrogen atom, a halogen atom, a C_{1-6} alkyl group or a group of the formula $-N(R^{25})R^{26}$; each of R^{25} and R^{26} which are independent of each other, is a hydrogen atom, a C_{1-4} alkyl group, a C_{1-4} alkoxy group, a C_{2-12} alkoxyalkyl group or a group of the formula $-SO_2R^{27}$; or R^{25} and R^{26}

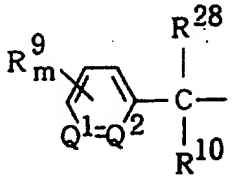
may form a 5- or 6-membered ring together with the nitrogen atom to which they are bonded; R^{27} is a C_{1-8} alkyl group or a phenyl group (which may be substituted by a halogen atom or a C_{1-4} alkyl group); each of Q^1 and Q^2 is a nitrogen atom or a group of the formula $-CR^9$; m is an integer of from 1 to 3; and n is 0, 1 or 2.

2. A benzylsulfide derivative of the formula (II):

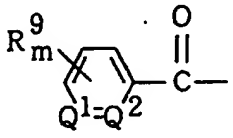


(II)

wherein R^1, R^2, R^3, R^4 and n are as defined in Claim 1; and B is an aralkyl or arylcarbonyl group of the formula (B1) or (B2):



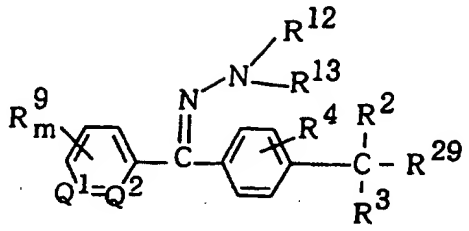
[B1]



[B2]

wherein R^9, R^{10}, m, Q^1 and Q^2 are as defined in Claim 1, and R^{28} is a halogen atom or a hydroxyl group.

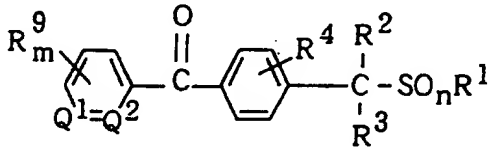
3. A benzophenonehydrazone derivative of the formula (III):



(III)

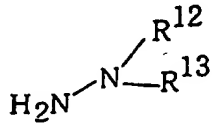
wherein R^4, R^9, R^{12}, R^{13}, m, Q^1 and Q^2 are as defined in Claim 1; each of R^2 and R^3 which are independent of each other, is a hydrogen atom or a C_{1-4} alkyl group, and R^{29} is a halogen atom, a mercapto group or a hydroxyl group.

4. A process for producing a benzylsulfide derivative wherein A is a group of the formula (A2) as defined in Claim 1, which comprises reacting a compound of the formula (IV):



(IV)

wherein R^1, R^2, R^3, R^4, R^9, m, n, Q^1 and Q^2 are as defined in Claim 1, with a compound of the formula (V1):

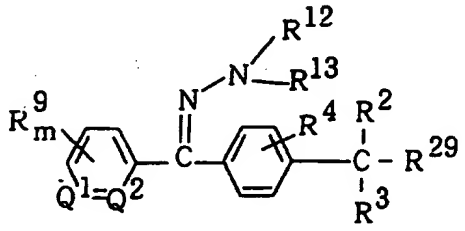


(V1)

wherein R^{12} and R^{13} are as defined in Claim 1.

5. A process for producing a benzylsulfide derivative

wherein A is a group of the formula (A2) as defined in Claim 1, which comprises reacting a compound of the formula (III);



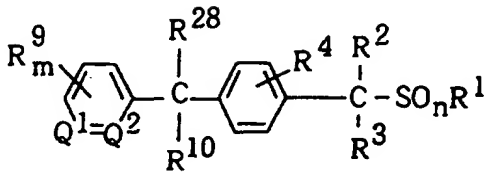
(III)

wherein R^2, R^3, R^4, R^9, R^{12}, R^{13}, R^{29}, m, Q^1 and Q^2 are as defined in Claim 3, with a compound of the formula (V2):

$$Z\text{-}R^1 \qquad (V2)$$

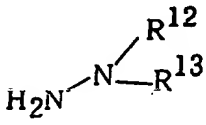
wherein Z is a halogen atom, a C_{1-4} alkylsulfonyl group or a benzenesulfonyloxy group (which may be substituted by a methyl group) when R^{29} is a mercapto group, or a group of the formula a $-S(O)_nM$ when R^{29} is a halogen atom, or a group of the formula $-SSR^1$ when R^{29} is a hydroxyl group; R^1 is a C_{1-6} alkyl group, a C_{1-4} cyanoalkyl group, a C_{3-6} cycloalkyl group, a C_{1-6} haloalkyl group, a C_{2-4} alkenyl group or a benzyl group (which may be substituted by a halogen atom); M is an alkali metal; and n is 0 or 2.

6. A process for producing a benzylsulfide derivative wherein A is a group of the formula (A1) as defined in Claim 1, which comprises reacting a compound of the formula (VI):



(VI)

wherein R^1, R^2, R^3, R^4, R^9, R^{10}, m, n, Q^1 and Q^2 are as defined in Claim 1, and R^{28} is a halogen atom, with a compound of the formula (Vl):



(Vl)

wherein R^{12} and R^{13} are as defined in Claim 1.

7. A pesticide containing a benzylsulfide derivative as defined in Claim 1, as an active ingredient.

INTERNATIONAL SEARCH REPORT

Inter nal Application No
PC:/JP 96/01055

A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C07C317/28 C07C323/29 C07D213/42 C07D213/53 C07C317/24
C07C317/18 C07C323/22 C07C323/16 C07D213/50 C07D213/32
C07C251/86 C07C319/00 C07C315/00 A01N33/26 A01N43/40

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 C07C C07D A01N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A,P	EP,A,0 662 472 (CIBA GEIGY AG) 12 July 1995 ---	1-7
A	EP,A,0 581 725 (CIBA GEIGY AG) 2 February 1994 ---	1-7
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Date of the actual completion of the international search	Date of mailing of the international search report
10 July 1996	-7.08.96

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Janus, S

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INTERNATIONAL SEARCH REPORT

Inv onal Application No
PCT/JP 96/01055

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 A01N41/04

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

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Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No
PCT/JP 96/01055

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		JP-A-	7242618	19-09-95
		ZA-A-	9500028	13-07-95
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		RU-C-	2002732	15-11-93
		US-A-	4980373	25-12-90

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(30) Priority Data:
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(71) Applicant *(for all designated States except US)*: CANCER RESEARCH CAMPAIGN TECHNOLOGY LIMITED [GB/GB]; Cambridge House, 6-10 Cambridge Terrace, Regent's Park, London NW1 4JL (GB).

(72) Inventors; and
(75) Inventors/Applicants *(for US only)*: CONNORS, Thomas [GB/GB]; MRC Toxicology Unit, MRC Laboratories, Woodmansterne Road, Carshalton, Surrey SM5 4EF (GB). KNOX, Richard [GB/GB]; The Institute of Cancer Research, Royal Cancer Hospital, 15 Cotswold Road, Belmont, Sutton, Surrey, SM2 5NG (GB). SHERWOOD, Roger [GB/GB]; PHLS Centre for Applied Microbiology & Research, Division of Biotechnology, Porton Down, Salisbrury, Wiltshire SP4 0JG (GB).

(74) Agents: GOLDIN, Douglas, Michael et al.; J.A. Kemp & Co., 14 South Square, Gray's Inn, London WC1R 5LX (GB).

(81) Designated States: AU, CA, JP, NZ, US, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).

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(88) Date of publication of the international search report:
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(54) Title: IMPROVEMENTS RELATING TO CANCER THERAPY

(57) Abstract

The system of the invention comprises: (i) a viral vector comprising a nucleotide sequence encoding a nitroreductase, which nitroreductase in capable of converting a prodrug into a cytotoxic drug; and (ii) a prodrug capable of being converted into a cytotoxic drug by the nitroreductase encoded by the vector.

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GA	Gabon				

INTERNATIONAL SEARCH REPORT

I ıational Application No
PCT/GB 94/02423

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 C12N15/53 C12N15/86 A61K48/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 A61K C12N

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Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP,A,0 330 432 (THE DEPARTMENT OF MOLECULAR PHARMACOLOGY, INSTITUTE OF CANCER RESEARCH) 30 August 1989 see column 3 and 7 ---	12
X	EP,A,0 540 263 (CANCER RESEARCH CAMPAIGN TECHNOLOGY LIMITED) 5 May 1993 see pages 4-6	12
Y	see the whole document --- -/--	1-11, 13-17

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Date of the actual completion of the international search	Date of mailing of the international search report
27 April 1995	19.05.95

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax (+31-70) 340-3016	Alt, G

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	BIOCHEM.PHARMACOL., vol.44, no.12, 15 December 1992, ENGLAND pages 2289 - 2295 ANLEZARK, G.M. ET AL. 'The bioactivation of 5-(aziridin-1-yl)-2,4-dinitrobenzamide (CB1954)--I. Purification and properties of a nitroreductase enzyme from Escherichia coli -- a potential enzyme for antibody-directed enzyme prodrug therapy (ADEPT)' see Figure 1	12
Y	see whole document	1-11, 13-17

Y	PROC.NATL.ACAD.SCI., vol.88, September 1991, USA pages 8039 - 8043 HUBER, B.E. ET AL. 'Retroviral-mediated gene therapy for the treatment of hepatocellular carcinoma: An innovative approach for cancer therapy' see page 8040, left-hand column, lines 6-25 to right-hand column, lines 1-14; page 8043, right-hand column, last paragraph	1-11, 13-17

Y	PROC.NATL.ACAD.SCI., vol.89, 1992, USA pages 33 - 37 MULLEN, C.A. ET AL. 'Transfer of the bacterial gene for cytosine deaminase to mammalian cells confers lethal sensitivity to 5-fluorocytosine: a negative selection system'	1-11, 13-17

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No
PCT/GB 94/02423

Patent document cited in search report	Publication date	Patent family member(s)		Publication date
EP-A-0330432	30-08-89	AU-A-	3439889	06-09-89
		EP-A-	0429454	05-06-91
		WO-A-	8907592	24-08-89
		JP-T-	3503761	22-08-91
EP-A-0540263	05-05-93	AU-A-	2776992	21-05-93
		EP-A-	0638123	15-02-95
		WO-A-	9308288	29-04-93
		JP-T-	7501692	23-02-95